Public Citizen 30 YEARS
Protecting Health, Safety & Democracy

Buyers Up • Congress Watch • Critical Mass • Global Trade Watch • Health Research Group • Litigation Group
Joan Claybrook, President

May 8, 2001

Dockets Management Branch
Food and Drug Administration
5630 Fishers Lane, Room 1061 (HFA-305)
Rockville, MD 20852

Re: Docket No. 94F-0008 — "Irradiation in the Production, Processing and Handling of Food"

To whom it may concern:

Under the provisions of 21 CFR §12.20-28, Public Citizen is requesting a formal evidentiary public hearing for the purposes of revoking the Food and Drug Administration's Final Rule on Docket No. 94F-0008 — "Irradiation in the Production, Processing and Handling of Food."[1]

Federal regulations require the FDA to demonstrate, before approving a food additive for human consumption, that the additive is "safe," which 21 CFR §170.3(i) defines as follows: "there is reasonable certainty in the minds of competent scientists that the substance is not harmful under the intended conditions of use."[2]

The FDA did not meet this requirement when issuing its Final Rule for this docket.

We have identified and seek to present at a public hearing genuine and substantial issues containing evidence that raises material issues of fact, and which questions in a material way the rationale of this ruling. Due to substantial, material shortcomings in the rationale of this ruling, potential risks to public health have not be sufficiently examined.

This ruling, which regards the use of X-rays to inspect cargo containers that may contain food, relies on three articles (References 1, 2, 3) to support the agency's conclusion that "no detectable radioactivity will be induced in food when an X-ray energy of 10 MeV and a dose of 0.5 Gy are not exceeded."[3] The use of these three articles to draw this conclusion is substantially flawed:

(1) Reference 1, a 1990 article published in the *Bulletin of the World Health Organization*,[4] states that "no detectable radioactivity will be induced in foodstuffs when an [X-ray] energy level of 10 MeV and a dose of 0.5 Gy are not exceeded." However, this statement was based on an "extrapolat[ion]" of "theoretical and experimental" studies that the article neither specifically quotes nor references.

Public Citizen is requesting a formal evidentiary public hearing on this matter.

94F-0008

Ralph Nader, Founder
215 Pennsylvania Ave SE • Washington, DC 20003 • (202) 546-4996 • www.citizen.org

1 HER 1

(2) Reference 2, Wakeford and Blackburn (1991),[5] is cited in the Final Rule to support the statement that electrons with energies of 8 MeV - 10 MeV induced an "extremely small level of radioactivity."[6] This statement is irrelevant to the petition, which regards the use of X-rays. Additionally, the Final Rule's statement that the "FDA would not expect any detectable radioactivity above background in food resulting from the petitioned use"[7] is based on no data or evidence whatsoever.

Furthermore, with regard to X-rays, Wakeford and Blackburn states that "X-rays of energy greater than 3 MeV could induce radioactivity." The study also states that "four isotopes can be activated at [X-ray] energies below 5 MeV and all have stable products; however, the reactions generate neutrons which could lead to indirect activation." Among the four isotopes are carbon-13, which has photoneutron activation threshold of 4.95 MeV; oxygen-17, which has a threshold of 4.2 MeV; and deuterium, which has a threshold of 2.2 MeV. Of deuterium, the study states: "at any photon energy in excess of 2.2 MeV, there will be some slight neutron induced activity in real food."[8]

Public Citizen is requesting a formal evidentiary public hearing on this matter.

(3) Reference 3, Findlay et al (1992),[9] which regards the induced radioactivity in food caused by electron beams, is irrelevant to the petition, which regards the use of X-rays.

Public Citizen is requesting a formal evidentiary public hearing on this matter.

The inappropriateness of using studies on electron beam irradiation to support conclusions about X-ray irradiation is reflected in a 1995 report by the International Consultative Group on Food Irradiation (ICGFI). The study states that "the neutron activity produced by 5 MeV X-rays is in the order of 60 times greater than that produced by 10 MeV electrons."[10] The study lists several radioactive isotopes that can be formed in food as a result of X-ray irradiation at energy levels below 5 MeV, including:

- carbon-14 (half-life 5,730 years);
- chlorine-36 (half-life 310,000 years);
- chlorine-38 (half-life 37.3 minutes);
- potassium-40 (half-life 1.28 billion years);
- potassium-42 (half-life 12.4 hours);
- phosphorus-32 (half-life 14.3 days);
- sodium-24 (half-life 15 hours); and
- sulfur-35 (half-life 88 days).[11]

The ICGFI report concludes that "increasing the energy of X-rays above 7.5 MeV would result in ... possible induction of radioactivity in the irradiated food."[12]

(The ICGFI report states that 10 MeV X-rays with a dose of 0.5 Gy "would not produce any significant radioactivity,"[13] but, like the 1990 article from the *Bulletin of the World Health Organization* referenced in the Final Rule, the ICGFI report neither quotes nor references any specific studies to support this statement.)

Taken together, these flaws in the FDA's Final Rule represent genuine and substantial issues containing evidence that raises material issues of fact and questions in a material way the rationale of the ruling. Due to these substantial, material flaws, potential risks to public health have not be sufficiently examined.

We request that a formal evidentiary public hearing on these issues be held at the earliest possible date.

Respectfully submitted,

Wenonah Hauter
Director,
Critical Mass Energy and Environment Program

Enclosures

cc: Dr. Bernard Schwetz
Mr. Joseph Levitt
Dr. Alan Rulis
Dr. Laura Tarantino

Notes

[1] 66 FR 18538, April 10, 2001.

[2] 21 CFR §170.3(i)

[3] 66 FR 18538, April 10, 2001.

[4] "Food safety aspects relating to the application of X-ray surveillance equipment: Memorandum from a WHO meeting." *Bulletin of the World Health Organization*, 68:297-301, 1990.

[5] Wakeford C.A. and Blackburn R. "Induction and detection of radioactivity in foodstuffs irradiated with 10 MeV electrons and X-rays." *Radiation Physics and Chemistry*, 38:29-38, 1991.

[6] 66 FR 18538, April 10, 2001.

[7] Ibid.

[8] Wakeford C.A. and Blackburn R. op cit.

[9] Findlay, D.J.S. et al. "Experimental electron beam irradiation of food and the induction of radioactivity." *Applied Radiation and Isotopes*, 43:567-575, 1992.

[10] "The development of X-ray machines for food irradiation (Proceedings of a consultants' meeting)." Vienna: International Consultative Group on Food Irradiation, Oct. 16-18, 1995.

[11] Ibid.

[12] Ibid.

[13] Ibid.

FDC date	State	City	Airport	FDC No.	Subject
03/22/01	TX	ROBSTOWN	NUECES COUNTY	1/2866	VOR/DME–A, AMDT 3
03/22/01	TX	ROBSTOWN	NUECES COUNTY	1/2867	GPS RWY 12, ORIG–B
03/22/01	UT	SALT LAKE CITY	SALT LAKE CITY INTL	1/2870	RNAV (GPS) RWY 35, ORIG
03/22/01	PA	PITTSBURGH	PITTSBURGH INTL	1/2877	2
03/22/01	NE	HASTINGS	HASTINGS MUNI	1/2882	GPS RWY 14, ORIG–B
03/22/01	OK	EL RENO	EL RENO MUNI AIR PARK	1/2894	VOR/DME RWY 35, AMDT 1A
03/23/01	FL	KEY WEST	KEY WEST INTL	1/2898	RADAR–1, AMDT 4A
03/23/01	FL	KEY WEST	KEY WEST INTL	1/2899	GPS RWY 9, ORIG–A
03/23/01	FL	KEY WEST	KEYWEST INTL	1/2900	GPS RWY 27, ORIG–A NDB OR GPS–A, AMDT
03/23/01	FL	KEYWEST	KEYWEST INTL	1/2902	15A
03/23/01	OK	NORMAN	UNIVERSITY OF OKLAHOMA WESTHEIMER.	1/2918	LOC RWY 3, AMDT 3C
03/23/01	OK	NORMAN	UNIVERSITY OF OKLAHOMA WESTHEIMER.	1/2919	VOR/DME RNAV RWY 3, ORIG–D
03/26/01	FL	ORMOND BEACH	ORMOND BEACH MUNI	1/2974	VOR OR GPS RWY 17, AMDT 1B
03/26/01	FL	ORMOND BEACH	ORMOND BEACH MUNI	1/2976	GPS RWY 8, ORIG
03/26/01	FL	ORMOND BEACH	ORMOND BEACH MUNI	1/2977	RADAR–1, AMDT 2B
03/26/01	WA	SPOKANE	SPOKANE INTL	1/2992	RNAV (GPS) RWY 3, ORIG GPS RWY 28, ORIG–B
03/26/01	NM	SANTA FE	SANTA FE MUNI	1/2993	(Replaces FDC 1/2274)
		NEW SMYRNA	NEW SMYRNA		
03/27/01	FL	BEACH	BEACH MUNI	1/3020	AMDT 1
		NEW SMYRNA	NEW SMYRNA		
03/27/01	FL	BEACH	BEACH MUNI	1/3023	RADAR–1, AMDT 3
03/27/01	AK	NOME	NOME	1/3025	GPS RWY 27, ORIG–B
03/27/01	AK	NOME	NOME	1/3026	GPS RWY 9, ORIG–B
03/27/01	AK	NOME	NOME	1/3027	GPS RWY 2, ORIG–B
			ASPEN–PITKIN COUNTY/ SARDY.		
03/27/01	CO	ASPEN	FIELD	1/3034	VOR/DME OR GPS–C AMDT 4B
03/27/01	GA	SAVANNAH	SAVANNAH INTL	1/3037	VOR OR TACAN OR GPS RWY 27, AMDT 15C
03/27/01	GA	SAVANNAH	SAVANNAH INTL	1/3038	ILS RWY 9, AMDT 25D
03/27/01	GA	SAVANNAH	SAVANNAH INTL	1/3039	HI–ILS RWY 9, AMDT 5
03/27/01	GA	SAVANNAH	SAVANNAH INTL	1/3040	HI–TACAN RWY 27, AMDT 3

[FR Doc. 01–8715 Filed 4–9–01; 8:45 am]
BILLING CODE 4910–13–M

DEPARTMENT OF HEALTH AND HUMAN SERVICES

Food and Drug Administration

21 CFR Part 179

[Docket No. 94F–0008]

Irradiation in the Production, Processing and Handling of Food

AGENCY: Food and Drug Administration, HHS.

ACTION: Final rule.

SUMMARY: The Food and Drug Administration (FDA) is amending the food additive regulations to provide for the safe use of a machine source of high energy x-rays to inspect cargo containers that may contain food. This action is in response to a petition filed by Analytical Systems Engineering Corp. (ASEC).

DATES: This rule is effective April 10, 2001. Submit written objections and request for a hearing by May 10, 2001.

ADDRESSES: Submit written objections to the Dockets Management Branch (HFA–305), Food and Drug Administration, 5630 Fishers Lane, rm. 1061, Rockville, MD 20852.

FOR FURTHER INFORMATION CONTACT: Andrew J. Zajac, Center for Food Safety and Applied Nutrition (HFS–206), Food and Drug Administration, 200 C St. SW., Washington, DC 20204, 202–418–3095.

SUPPLEMENTARY INFORMATION:

I. Introduction

In a notice published in the **Federal Register** of February 24, 1994 (59 FR 8995), FDA announced that a food additive petition (FAP 4M4407) had been filed by Analytical Systems Engineering Corp., 5400 Shawnee Rd., suite 100, Alexandria, VA 22312. The petition proposed that the food additive regulations in § 179.21 *Sources of radiation used for inspection of food, for inspection of packaged food, and for controlling food processing* (21 CFR 179.21) be amended to provide for the safe use of a machine source of high energy x-rays to inspect cargo containers that may contain food. In a letter dated October 12, 2000, ASEC (now ACS Defense, Inc., 2001 North Beauregard St., Alexandria, VA 22311) informed FDA of the transfer of their rights to FAP 4M4407 to R. F. Reiter and Associates, 850 Oak Chase Circle, Fairfax Station, VA 22039.

II. Evaluation of Safety

A source of radiation used for the purpose of inspection of foods meets the definition of a food additive under section 201(s) of the Federal Food, Drug, and Cosmetic Act (the act) (21 U.S.C. 321(s)). Under section 409(c)(3)(A) of the act (21 U.S.C. 348(c)(3)(A)), a food additive cannot be approved for a particular use unless a fair evaluation of the data available to FDA establishes that the additive is safe for that use. FDA's food additive regulations in § 170.3(i) (21 CFR 170.3(i)) define safe as "a reasonable certainty in the minds of competent scientists that the substance is not harmful under the intended conditions of use."

III. Evaluation of the Safety of the Petitioned Use of a Source of Radiation

Machine sources that produce high energy x-rays may be used to screen large cargo containers for illegal drugs and other contraband. To be able to penetrate large cargo containers, these x-ray systems need to operate with x-ray energies higher than those used for screening smaller articles (Ref. 1). The present petition proposes the use of x-rays produced by an electron linear accelerator operating at energy levels of up to 10 million electron volts (MeV) to inspect large cargo containers that may contain food, provided that the maximum dose absorbed by the food does not exceed 0.5 gray (0.5 Gy). Because the probability of inducing a change in the nucleus of an atom absorbing x-rays increases with the energy of the x-ray, the potential for induced radioactivity in the finished foodstuff needs to be assessed. Current regulations authorize the use of x-rays at energies up to 0.5 MeV to inspect cargo, including food, provided the absorbed dose does not exceed 10 Gy (§ 179.21). This petition seeks to raise the energy limit for x-rays from 0.5 MeV to 10 MeV, however, the petition also proposes to limit the maximum absorbed dose to 0.5 Gy, well below the 10 Gy level previously established as safe for food inspection. Accordingly, FDA has concluded that there is no need to evaluate changes in the food subjected to x-rays other than the potential for induced radioactivity.

The petitioner submitted a number of published articles and other study reports containing data and information on the induction of radioactivity in food. One of the reports that the petitioner relied on to demonstrate that the petitioned use of the source of radiation is safe is from the World Health Organization (WHO). This WHO report concluded that no detectable radioactivity will be induced in foodstuffs by x-rays with a maximum energy level of 10 MeV when a dose of 0.5 Gy is not exceeded (Ref. 1).

As part of FDA's safety review of the petition, the agency evaluated two studies in which various foods were irradiated with either x-rays or electron beams at energies sufficient to induce radioactivity. Radioactivity is the result of changes in the nucleus of an atom induced, for example, by interaction with x-rays. Because the elemental composition of the foods that were studied is representative of foods in general, the results of the two studies may reasonably be applied to other foods subjected to these test conditions. In one study, three types of food were irradiated with high energy bremsstrahlung[1] produced by an electron linear accelerator that generated predominately 8 MeV electrons (approximately 7 percent of the electrons were in the range of 8 to 10 MeV and less than 2 percent were in the range of 10 to 12 MeV) (Ref. 2). The types of food that were irradiated were codfish, rice, and a macerated meat product. These foods received doses ranging from 8.8 to 14 kiloGy (kGy) (17,600 to 28,000 times higher than the maximum petitioned dose level of 0.5 Gy). The authors concluded that the induced activities in the foods that were observed immediately after irradiation are approximately the same as natural background levels, and that any induced activities drop quickly. According to the data presented in the paper, by 1 day after irradiation, induced levels of radioactivity were typically about 10 percent of those initially observed. Because of the extremely small level of radioactivity that was induced in foods after receiving doses thousands of times higher than the maximum petitioned dose, FDA would not expect any detectable radioactivity above background in food resulting from the petitioned use of the source of radiation at doses up to 0.5 Gy.

In the second study, samples of chicken, prawns, cheeses, and spices were irradiated with electron beams at energies of 10 MeV and 20 MeV and induced radioactivity was measured (Ref. 3). In this study, the mechanisms responsible for the induced radioactivity in the samples were photonuclear reactions induced by bremsstrahlung and electronuclear reactions induced by the electron beams. The authors noted that when food is irradiated with electron beams with an energy at or below 10 MeV, the induced radioactivity in food is essentially zero. Therefore, to produce measurable radioactivity in food, irradiations were also carried out at 20 MeV. The authors stated that the study with 20 MeV irradiations was intended to simulate a gross malfunction of an electron beam irradiation plant. The authors concluded that, as expected, no measurable radioactivity induced at 10 MeV was detected, and that even at energies as high as 20 MeV and doses up to 10 kGy (i.e., 20,000 times the maximum petitioned dose level of 0.5 Gy), the specific activity after 1 day was approximately 0.01 Becquerel/gram (Bq/g), which is negligible (Ref. 3).

[1] Bremsstrahlung refers to the type of radiation which is emitted when high-speed electrons are suddenly decelerated due to interactions with atomic nuclei.

IV. Conclusion of Safety

FDA has evaluated the data submitted in the petition and other relevant material and concludes that no detectable radioactivity will be induced in food when an x-ray energy of 10 MeV and a dose of 0.5 Gy are not exceeded. Therefore, the agency concludes that the proposed use of x-radiation, produced by a machine source at energies of 10 MeV or lower, to inspect food, is safe and that the conditions listed in §179.21 should be amended as set forth below.

In accordance with § 171.1(h) (21 CFR 171.1(h)), the petition and the documents that FDA considered and relied upon in reaching its decision to approve the petition are available for inspection at the Center for Food Safety and Applied Nutrition by appointment with the information contact person listed above. As provided in § 171.1(h), the agency will delete from the documents any materials that are not available for public disclosure before making the documents available for inspection.

V. Environmental Impact

In the notice of filing, FDA gave interested parties an opportunity to submit comments on the petitioner's environmental assessment. FDA received no comments in response to that notice. The agency has carefully considered the potential environmental effects of this action. FDA has concluded that this action will not have a significant impact on the human environment, and that an environmental impact statement is not required. The agency's finding of no significant impact and the evidence supporting that finding, contained in an environmental assessment, may be seen in the Dockets Management Branch (address above) between 9 a.m. and 4 p.m., Monday through Friday.

VI. Paper Reduction Act of 1995

This final rule contains no collection of information. Therefore, clearance by the Office of Management and Budget under the Paperwork Reduction Act of 1995 is not required.

VII. Objections

Any person who will be adversely affected by this regulation may at any time file with the Dockets Management Branch (address above) written objections by May 10, 2001. Each objection shall be separately numbered, and each numbered objection shall specify with particularity the provisions of the regulation to which objection is made and the grounds for the objection. Each numbered objection on which a hearing is requested shall specifically so

state. Failure to request a hearing for any particular objection shall constitute a waiver of the right to a hearing on that objection. Each numbered objection for which a hearing is requested shall include a detailed description and analysis of the specific factual information intended to be presented in support of the objection in the event that a hearing is held. Failure to include such a description and analysis for any particular objection shall constitute a waiver of the right to a hearing on the objection. Three copies of all documents are to be submitted and are to be identified with the docket number found in brackets in the heading of this document. Any objections received in response to the regulation may be seen in the Dockets Management Branch between 9 a.m. and 4 p.m., Monday through Friday.

VIII. References

The following references have been placed on display in the Dockets Management Branch (address above) and may be seen by interested persons between 9 a.m. and 4 p.m., Monday through Friday.

1. WHO, "Food safety aspects relating to the application of X-ray surveillance equipment: Memorandum from a WHO meeting," Bulletin of the World Health Organization, vol. 31, pp. 297–301, 1990.

2. Wakeford, C. A. and R. Blackburn, "Induction and Detection of Radioactivity in Foodstuffs Irradiated with 10 MeV Electrons and X-rays," *Radiation Physics and Chemistry*, vol. 38, No. 1, pp. 29–38, 1991.

3. Findley, D. J. S., T.V. Parson, and M. R. Sene, "Experimental Electron Beam Irradiation of Food and the Induction of Radioactivity," *Applied Radiation and Isotopes*, vol. 43, pp. 567–575, 1992.

List of Subjects in 21 CFR Part 179

Food additives, Food labeling, Food packaging, Radiation protection, Reporting and recordkeeping requirements, Signs and symbols.

Therefore, under the Federal Food, Drug, and Cosmetic Act and under authority delegated to the Commissioner of Food and Drugs and redelegated to the Director, Center for Food Safety and Applied Nutrition, 21 CFR part 179 is amended as follows:

PART 179—IRRADIATION IN THE PRODUCTION, PROCESSING AND HANDLING OF FOOD

1. The authority citation for 21 CFR part 179 continues to read as follows:

Authority: 21 U.S.C. 321, 342, 343, 348, 373, 374.

2. Section 179.21 is amended by adding paragraphs (a)(4), (b)(1)(iii), and (b)(2)(iv) to read as follows:

§ 179.21 Sources of radiation used for inspection of food, for inspection of packaged food, and for controlling food processing.

* * * * *

(a) * * *

(4) Machine sources producing X-radiation at energies no greater than 10 million electron volts (MeV).

(b) * * *

(1) * * *

(iii) The maximum energy of X-radiation emitted by machine source.

(2) * * *

(iv) A statement that no food shall be exposed to a radiation source listed in paragraph (a)(4) of this section so as to receive a dose in excess of 0.5 gray (Gy).

Dated: April 3, 2001.

L. Robert Lake,

Director of Regulations and Policy, Center for Food Safety and Applied Nutrition.

[FR Doc. 01–8755 Filed 4–9–01; 8:45 am]

BILLING CODE 4160–01–S

DEPARTMENT OF HEALTH AND HUMAN SERVICES

Food and Drug Administration

21 CFR Part 579

[Docket No. 99F–2799]

Irradiation in the Production, Processing, and Handling of Animal Feed and Pet Food; Irradiation

AGENCY: Food and Drug Administration, HHS.

ACTION: Final rule.

SUMMARY: The Food and Drug Administration (FDA) is amending the food additive regulations to reflect approval of a food additive petition (FAP) filed by Sterigenics International, Inc. (now IBA Food Safety Division) that provides for irradiation of various animal feeds and feed ingredients for microbial control.

DATES: This rule is effective April 10, 2001. Submit written objections and request for a hearing by May 10, 2001.

ADDRESSES: Submit written objections to the Dockets Management Branch (HFA–305), Food and Drug Administration, 5630 Fishers Lane, rm. 1061, Rockville, MD 20852.

FOR FURTHER INFORMATION CONTACT: John D. McCurdy, Center for Veterinary Medicine (HFV–222), Food and Drug Administration, 7500 Standish Pl., Rockville, MD 20855, 301–827–0171.

SUPPLEMENTARY INFORMATION: In a notice published in the **Federal Register** of September 3, 1999 (64 FR 48409), FDA announced that a food additive petition (FAP 2243) had been filed by SteriGenics International, Inc., 4020 Clipper Ct., Fremont, CA 94538–6540. The petition proposed to amend the food additive regulations in part 21 CFR part 579 Irradiation in the Production, Processing, and Handling of Animal Feed and Pet Food to provide for the irradiation of various animal feeds and feed ingredients to control microbial contaminants. The notice of filing provided for a 60-day comment period. The agency received no comments.

FDA has evaluated data submitted by the sponsor of the petition and concludes that the data establish the safety and functionality of irradiation for use as proposed.

This final rule extends the ability to irradiate all animal feeds for the purpose of microbial disinfection, therefore, references to laboratory animals have been deleted from the regulation. Also, paragraph (b)(2) has been added to § 579.22 to make clear that as long as an irradiated feed ingredient is less than 5 percent of the final product, the final product may be irradiated without conflicting with the statement in § 579.22(b)(1) that the ionizing radiation is used or intended for use in single treatment.

In accordance with § 571.1(h) (21 CFR 571.1(h)), the petition and the documents that FDA considered and relied upon in reaching its decision to approve the petition are available for inspection at the Center for Veterinary Medicine by appointment with the information contact person listed above. As provided in § 571.1(h), the agency will delete from the documents any materials that are not available for public disclosure before making the documents available for inspection.

FDA has determined under 21 CFR 25.32(j) that this action is of type that does not individually or cumulatively have a significant effect on the human environment. Therefore, neither an environmental assessment nor an environmental impact statement is required.

Any person who will be adversely affected by this regulation may at any time file with the Dockets Management Branch (address above) written objections by May 10, 2001. Each objection shall be separately numbered, and each numbered objection shall specify with particularity the provisions of the regulation to which objection is made and the grounds for the objection. Each numbered objection on which a hearing is requested shall specifically so state. Failure to request a hearing for any particular objection shall constitute a waiver of the right to a hearing on that objection. Each numbered objection for

derson A. (1988)
hanol C_2H_5OH,
Appl. Phys. **26**,

istry of organic
by Farhataziz I.

Yamauchi S. and Kanno H. (1989) Observation of resolved Raman bands for solvated ions in glassy alcohol solutions. *Chem. Phys. Lett.* **154**, 248.

Zietlow J. P., Cleveland F. F. and Meister A. G. (1950) Substituted methanes. III. Raman spectra, assignment and force constants for some trichloromethanes. *J. chem. Phys.* **18**, 1076.

Radiat. Phys. Chem. Vol. 38, No. 1, pp. 29–38, 1991
Int. J. Radiat. Appl. Instrum., Part C
Printed in Great Britain.

0146-5724/91 $3.00 + 0.00

INDUCTION AND DETECTION OF RADIOACTIVITY IN FOODSTUFFS IRRADIATED WITH 10 MeV ELECTRONS AND X-RAYS

C. A. WAKEFORD and R. BLACKBURN†
Department of Chemistry and Applied Chemistry, University of Salford, Salford M5 4WT, England

A. J. SWALLOW
Paterson Institute for Cancer Research, Christie Hospital and Holt Radium Institute, Manchester M20 9BX, England

(Received 3 April 1990; received for publication 27 September 1990)

Abstract—Induction of radioactivity by direct (γn) and indirect (nγ) means was measured in solutions and foods irradiated with 10 MeV electrons and X-rays, to a dose of 20 kGy. Both types of reaction occur to a very limited extent following electron irradiation and this is attributed to the low efficiency of bremsstrahlung production in the target. For X-rays below the 1981 JECFI recommended 5 MeV upper limit of energy, only four (γn) reactions are possible and all have stable products. However, the neutrons generated can undergo moderation and capture by a variety of nuclides. Of particular interest is the reaction $^2H(\gamma n)^1H$ since this has an abnormally low (γn) threshold of 2.2 MeV and deuterium occurs with a natural abundance of 0.015%; hence some neutron production in food is inevitable. A neutron flux of 1.79×10^5 [n cm^{-2} s^{-1}] [μg(D) cm^{-3}]$^{-1}$[kGy s^{-1}]$^{-1}$ was estimated from the activity induced in a variety of neutron monitors, especially manganese-55, as measured by gamma spectrometry and Cerenkov techniques. Induced activities at the end of irradiation calculated for beef using this flux value total 90 Bq kg^{-1} which is of the same order as natural levels in food (approx. 100 Bq kg^{-1}), and are in broad agreement with NRPB data.

INTRODUCTION

In accordance with international recommendations of a Joint FAO/WHO/IAEA Expert Committee (JECFI, 1981), the U.K. Advisory Committee on Irradiated and Novel Foods (ACINF, 1986) concluded that an overall average radiation dose of 10 kGy administered at energies of less than 5 MeV in the case of X- and γ-rays and 10 MeV in the case of electrons would not induce significant radioactivity in food. It was also thought unlikely that 10 MeV X- or γ-rays would induce significant radioactivity in practice. Since machine-produced radiations are increasingly available and are considered to offer economic and operational advantages over isotopic sources (Langunas-Solar and Matthews, 1985) we have now further examined the induction of radioactivity by electrons and X-rays with energies up to 10 MeV.

The ability to induce nuclear changes (and hence radioactivity) depends upon the energy of photons. These may be produced directly and deliberately as X-rays by allowing the electron beam of a machine source to fall upon a suitable target or they may arise indirectly from an electron beam impinging on some target material external to the electron source, such as the foodstuffs themselves, packaging or shielding and constructional materials associated with the radiation source or handling devices. Since the fractional conversion of energy is given by [E(MeV) $\times$ Z/800], the elements of low atomic number in foodstuffs themselves will effect little conversion to photons, but the other materials need to be considered.

Types of photon-induced nuclear reactions

Isomeric activation is a theoretical possibility. The photon is absorbed by the target nucleus, resulting in a metastable (isomeric) energy level of the nucleus which is usually very short-lived. Experimental studies (Glass and Smith, 1959; Miller and Jensen, 1987) and theoretical studies (Becker, 1983; Rogers, 1964) indicate that isomeric activation is insignificant in foods, although silver isomers could be produced (half-life 44 s) at activities of less than 1 Bq kg^{-1} (Tuchscheerer, 1966).

At high energies, absorption of a photon by the target nucleus can lead to the subsequent ejection of a particle, e.g. a neutron, proton or triton. At the photon energies likely to be relevant in food irradiation both Rogers and Becker conclude that the only photon-induced process of significance is photoneutron (γn) production. The high hydrogen content of foodstuffs makes the photoneutron

†To whom correspondence should be addressed.

reaction D(γn)H of special interest since deuterium occurs naturally in hydrogen to the extent of 0.015%. This reaction has an abnormally low threshold energy (2.2 MeV). The lowest threshold energy of any stable nuclide is 1.7 MeV (beryllium-9) so that there is no possibility of photoneutron production resulting from isotopic radiation sources. Between 1.7 and 5 MeV the only nuclei which undergo such (γn) reactions are deuterium, beryllium-9, carbon-13 and oxygen-17. With the exception of beryllium these nuclides are present as minor isotopes of common elements in food, and although they all undergo (γn) reactions which lead only to stable isotopes, they liberate neutrons which can subsequently undergo moderation and radiative capture reactions with a variety of elements, leading to radioactivity.

In addition direct (γn) activation of certain minor elements could occur at energies up to 10 MeV, e.g. $^{127}I(\gamma n)^{126}I$ for which the threshold is 9.1 MeV. Indirect activation by capture of photoneutrons can only be a second-order effect for electron irradiations of low atomic number materials, where conversion is low, but is more likely to be significant for X-irradiation.

Previous work and experimental strategy

Previous attempts to estimate the likely levels of induced radioactivity in foodstuffs have taken the form of either theoretical calculations, or experimental irradiation of representative foods (using isotopic sources, electron beam and/or X-radiation) and subsequent attempts at detection of radioactivity, usually by gamma spectrometry.

Rogers (1964) calculated the radioactivity arising from cobalt-60 γ-rays, and electron and X-ray irradiations (5–15 MeV) of food, and activation by (γp), (γn) and (nγ) reactions; he concluded that activation was insignificant for X-rays up to 5 MeV and electrons up to 7 MeV but considered that the use of higher electron energies would warrant further experimental study. Later, after considering both (γn) and (nγ) processes, it was concluded (Leboutet and Aucouturier, 1985) that radioactivity produced by 10 kGy irradiation with electrons of energy up to 10–11 MeV would be only a few per cent of natural levels, while X-rays of energy greater than 3 MeV could induce radioactivity at comparable levels; these activities would then decay within a few days. Becker (1983), on the other hand, calculated the induced activity from electron irradiation only, predicting that induced activity at 10 MeV would be insignificant. He concluded that (nγ) reactions were more significant than (γn) reactions in contributing to the activity at the end of the irradiation but were less important at longer times when food would probably be consumed; he therefore did not include calculations of the induced radioactivity resulting from (nγ) capture in his final analysis. In recent experimental work (Miller and Jensen, 1987) beef was irradiated to doses in the range 200–300 kGy with 10 and 13.5 MeV electrons, and also cobalt-60 γ-rays. No induced radioactivity of any sort was detected with γ-rays or 10 MeV electrons, even at these artificially high doses. Nitrogen-13 and sodium-24 were, however, detected by gamma spectrometry in beef irradiated by 13.5 MeV electrons. The threshold for the reaction $^{14}N(\gamma n)^{13}N$ is 10.6 MeV and the degree of activity obtained was in agreement with the above predictions of Becker. The activity of sodium-24 under these conditions was found to be 80 times that predicted by Becker, assuming that it was produced by the neutron capture reaction $^{23}Na(n\gamma)^{24}Na$. Miller and Jensen concluded that at 13.5 MeV, sources other than deuterium must be contributing to photoneutron production but were unable to identify them. We suggest that the photoneutrons generated by the (γn) reaction on nitrogen-14 could be responsible for the excess of sodium-24 activity, a view supported by the observation of nitrogen-13 activity. This reaction cannot occur at 10 MeV and the neutron flux at this energy would probably be insufficient to produce detectable levels of sodium-24.

In a similar study (Furuta *et al.*, 1990), very large doses of up to 100 kGy were used to maximize the probability of detecting any radioactivity induced in pepper irradiated by 10 MeV electrons either alone or after addition of one of several compounds containing elements whose (γn) thresholds are less than 10 MeV. Activity levels in spiked samples were used to estimate activity in samples of pepper alone, which showed no activity in excess of background. No recent experimental work using X-rays up to 5 MeV energy has been reported and it appears that the 5 MeV limit was suggested largely on the basis of theoretical considerations and inferences drawn from electron irradiations (Koch and Eisenhower, 1967).

Experimental studies involving irradiation of real foods containing natural levels of activatable isotopes suffer from the disadvantages that the size and nature of the samples are not always the most suitable for counting. Moreover, the levels of radioactivity induced are so low as to place undue emphasis on the reliability and reproducibility of background measurements, and on the stability of counting equipment. High resolution gamma spectroscopy offers the only viable direct counting method and offers obvious advantages for the identification of γ-emitting radioisotopes although it suffers from rather low counting efficiencies, especially at high photon energy. Cerenkov counting is considerably more efficient, in particular for those isotopes that have a high maximum beta energy, and hence can detect lower levels of activity. However, identification of nuclides is more difficult, and since a transparent medium is essential, it cannot therefore be directly applied to food materials.

In this study we have adopted a 'worst-case' strategy in which deliberate doping was used to increase the concentration of constituents likely to undergo (γn) and/or (nγ) activation and whose products are easily distinguished by their energies of emission (in the case of gamma spectroscopy) or half-lives (in the case of Cerenkov counting). The data so obtained is much less susceptible to background and ultimate sensitivity factors but can easily be extrapolated to calculate radioactivity induced in natural foods containing only normal concentrations of trace elements. In particular we have attempted to estimate the contribution of neutron activities, by measuring the neutron fluxes generated in foods and in monitor solutions. Of interest is the contribution made to indirect activation of food following photon interaction with oxygen-17, carbon-13, and, in particular, deuterium [which have (γn) thresholds less than 5 MeV]. Although our measurements are approximate they serve to provide an upper limit of induced radioactivity.

EXPERIMENTAL

1. Irradiation of aqueous solutions

Two solutions in deuterium oxide were used, one containing 0.1 M manganese sulphate and 0.1 M sodium iodide, the other 0.1 M sodium chloride. Manganese-55 and iodine-127 are both 100% abundant isotopes and have high neutron capture cross-sections in both the thermal and epithermal resonance region of the neutron energy spectrum (Garber and Kinsey, 1976). The products, manganese-56 and iodine-128, have half-lives of 2.58 h and 25 min respectively, short enough to show reasonable activity after short irradiations but long enough to facilitate detection and measurement by gamma spectrometry. Consequently they can be used as monitors for neutrons. The stable isotopes sodium-23 and chlorine-37 are activated to sodium-24 (half-life 15 h) and chlorine-38 (half-life 37 min) by the photoneutrons. Both these nuclides emit beta particles of high maximum energy (1.39 and 4.81 MeV respectively) which are efficiently counted by the Cerenkov technique. Iodine-127 has a (γn) threshold of 9.1 MeV and the resulting iodine-126 can be used to monitor direct activation. Control experiments were performed with solutions using light water so as to give an estimate of the activity induced by sources of neutrons other than deuterium, such as oxygen-17, carbon-13, iodine-127 and possibly the copper target.

Each solution (23 cm^3) was irradiated to a dose in the range 15–20 kGy by fast electrons and bremsstrahlung produced in a copper target.

2. Irradiation of whole foods

Three typical food materials, codfish, rice (which may be regarded as essentially having the composition of starch), and a macerated meat product (0.4% fibre, 2% ash, 7.5% protein and 4% oil) were irradiated directly by 0–10 MeV X-rays to a maximum dose of 15–20 kGy and subsequently analysed by gamma spectrometry. The deuterium content of some samples was artificially increased by addition of deuterium oxide. Manganese sulphate solution was used as a monitor for neutron production and was either intimately mixed with the foodstuffs or contained in discrete 5 cm^3 plastic vials distributed randomly throughout the sample. These monitor solutions were combined before examination for radioactivity and it was shown subsequently that there was excellent agreement between results obtained by both methods.

3. Constructional and shielding materials

While a 5 MeV X-ray beam would be below the threshold energy required to initiate (γn) reactions in the components of constructional and/or shielding materials, adventitious photons from a 10 MeV beam of electrons may generate photoneutrons and hence radioactivity in food. The production of photoneutrons in brass, concrete, steel and lead was therefore briefly examined.

The targets, of area 100 cm^2 and of thicknesses between 1 and 4 cm (sufficient to completely stop the electron beam), were bombarded to a dose of 20 kGy with 10 MeV electrons. Neutrons were monitored with a 0.1 M manganese sulphate solution, 575 cm^3 of which were contained in a plastic bottle located immediately behind the target, immersed in 4000 cm^3 of water in a plastic tank, and subtending a solid angle of 0.7 π. The resulting manganese-56 activity was measured by gamma spectrometry.

Irradiation conditions

Samples were irradiated at the Paterson Institute using a LINAC which generates electrons predominantly of energy 8 MeV. The leading edge of each pulse produced electrons of higher energy, with approx. 7% of the total being in the range 8–10 MeV and less than 2% in the range 10–12 MeV. For brevity we refer to these as 10 MeV electrons. This equipment could not be used to generate 5 MeV X-rays but could generate bremsstrahlung X-rays with a maximum energy in the region of 10 MeV, i.e. the energy likely to be produced, albeit inefficiently, by 10 MeV electrons. For the purposes of photoneutron production from deuterium the precise energy of the X-radiation is of little consequence since the cross-section of the D(γn)H reaction is not highly sensitive to photon energy, (IAEA, 1974). Although it achieves a maximum of 2.4 mbarns at 4.4 MeV it is 1.3 mbarns near threshold and at 10 MeV and therefore the maximum error introduced by the assumption of no variation of cross-section with energy is probably less than a factor of two.

For 10 MeV electron irradiations the solutions were contained in polythene vials (23 cm^3). These were of a size and shape such that the electron beam was uniform over the sample, located at 1 m from the accelerator exit window, and completely absorbed in the solution. Dosimetry was performed with Fricke solution.

For X-ray irradiations, bremsstrahlung (0–10 MeV) was generated by allowing the 10 MeV electron beam to impinge upon a water-cooled copper target (thickness 1 cm). The use of copper instead of the customary tungsten or tungsten alloy avoids the problem of neutron production in the target since copper isotopes have a high (γn) threshold while several tungsten isotopes lead to photoneutron production. The (γn) thresholds and isotopic abundances for ^{182}W, ^{183}W, ^{184}W and ^{186}W are respectively; (8.1 MeV, 26.4%), (6.2 MeV, 14.4%), (7.4 MeV, 30.6%) and (7.2 MeV, 28.4%), whilst those for ^{63}Cu and ^{64}Cu are respectively, (10.9 MeV, 69.1%) and (9.9 MeV, 30.9%). Samples were irradiated in a region of uniform photon flux at a distance of 17 cm. from the target. Once again Fricke dosimetry was used.

The activities induced in the irradiated samples and their associated neutron monitors were measured as quickly as possible (30–90 min) after irradiations of duration up to 30 min using X-rays and 1 min using electrons, counting being continued for as long as there was a significant signal.

Gamma spectrometry

A high resolution GeLi γ-ray detector (active volume 50 cm³; nominal resolution 2.1 KeV at 1.33 MeV; relative efficiency 3.3%) coupled with a Canberra Series-35 4000 channel multichannel analyser with automatic background correction was used to identify and quantify γ-ray photons emitted by irradiated samples. The counting efficiencies characterizing the different counting geometries presented by the samples to the detector were determined by dissolving an absolutely known activity of sodium-22 in water contained in vessels of appropriate shapes and sizes. Energy dependence of counting efficiency was measured by the standard europium-152 method.

Cerenkov counting

This technique was used to measure the activity of chlorine-38 and sodium-24, and occasionally manganese-56. Chlorine-38 and sodium-24 are counted only with very low efficiency by γ-ray counting but the high energy of their beta emissions gives Cerenkov efficiencies as high as 60 and 18% respectively. The Cerenkov characteristics of light and heavy water differ to a negligible extent. Polyethylene counting vials were used in conjunction with an Intertechnique SL30 scintillation counter. It is essential to transfer the liquids from the irradiation containers to new counting vials since the luminescence signal from irradiated vials enormously outweighs any signal due to radioactivity.

Calculation of neutron fluxes

Measured monitor activities were corrected back to their values at the end of the relevant irradiation period and the thermal neutron fluxes, ϕ, produced in the irradiated samples were calculated from the end-of-irradiation activity using the equation;

$$\Phi = \frac{(\text{Bq g}^{-1}\text{ monitor element}) \times (\text{At.wt of monitor})}{0.6\sigma[1-\exp(-\lambda t_{irr})]}\ \text{n cm}^{-2}\text{ s}^{-1}$$

where σ = thermal neutron elemental cross-section (barns) for activation, λ = decay constant (s^{-1}) of the activated monitor isotope, and t_{irr} = duration of irradiation in seconds.

The validity of the assumption that all the neutrons are moderated to thermal energies is discussed later.

Finally, the neutron fluxes generated in the irradiated material per unit concentration of deuterium, D, per unit dose rate were calculated in units of $[\text{n cm}^{-2}\text{ s}^{-1}][\mu\text{g(D) cm}^{-3}]^{-1}[\text{kGy s}^{-1}]^{-1}$ (Table 1). It is assumed that deuterium comprises 0.015% of all normal hydrogen. Dry rice is assumed to be chemically equivalent to starch, i.e. it has the empirical formula $[C_6H_{10}O_5]_4$, and both meat and fish are regarded essentially as water with regard to hydrogen content.

These neutron flux calculations take no account of fast neutrons leaking from our irradiated samples before they become thermalized. The degree to which this occurs depends not only upon the nature of the material but also upon the size and shape of the objects to be irradiated; large volumes of food could produce virtually complete thermalization and hence greater concomitant radioactivation due to the generally higher radiative capture cross-sections at thermal energies. Values obtained from the small experimental samples used in this study are likely to underestimate the activity generated in large samples. Although a precise correction would be difficult, we may attempt a semiquantitative correction using the relationship (Liverhant, 1960);

$$\text{Non-leakage fraction} = \tan^{-1}(B/\Sigma)/B/\Sigma,$$

where $\Sigma = (3L^2)^{-1/2}$.

The Fermi Age, L^2, is dependent only on the moderating power of the material, being 31.4 cm² for light water and 121 cm² for heavy water. The constant 'B' depends only on the size and shape of the sample. For a sphere $B^2 = (\pi/\text{radius})^2$ while for a cylinder $B^2 = (2.4/\text{radius})^2 + (\pi/\text{height})^2$. For a cylinder of the type used in the irradiation of meat the non-leakage fraction is 0.24. The slightly smaller vessels used for other foods have a fraction of about 0.15. For our experimental food irradiations the moderating ability of the mixtures is almost entirely due to the hydrogen content and is little affected by the presence of carbon or added deuterium, and was taken as being equivalent to that of water.

In our solution studies we may be underestimating the neutron production by more than a factor of 14 in light water and by as much as 25 in pure deuterium oxide; in the irradiated foods the factor is probably about 5.

RESULTS

1. Irradiation of aqueous solutions (Table 1)

(i) Fast electrons. Only traces of induced radioactivity (manganese-56, iodine-128 and iodine-126) were detected in the heavy water samples after irradiation to a dose of 20 kGy with 10 MeV electrons. However, the presence of iodine-126 activity clearly shows that the 10 MeV electron beam generates sufficient high energy (> 9.1 MeV) bremsstrahlung to initiate the photoneutron reaction $(^{127}I(\gamma n)^{126}I$. Production of iodine-126 by the alternative mechanism $^{127}I(n, 2n)^{126}I$ may be discounted on grounds of energy.

(ii) Bremsstrahlung. Irradiation with bremsstrahlung X-rays to a dose of 15 kGy of the same heavy water solutions produced easily measurable levels of manganese-56, iodine-128, chlorine-38 and sodium-24. These result from the capture reactions;

$$^{55}Mn(n\gamma)^{56}Mn, ^{127}I(n\gamma)^{128}I,$$

$$^{37}Cl(n\gamma)^{38}Cl, ^{23}Na(n\gamma)^{24}Na.$$

Significant manganese-56 activity was also observed in light water solutions. The neutron flux per unit deuterium concentration per unit dose rate in light water was found to be higher than that calculated from activity measurements in heavy water, indicating that there are significant sources of neutrons other than D(γn)H.

Neutron fluxes calculated from activities of manganese-56, sodium-24 and chlorine-38 are in reasonable agreement, giving a mean value of $325[\text{n cm}^{-2}\text{ s}^{-1}][\mu\text{g(D) cm}^{-3}]^{-1}[\text{kGy s}^{-1}]^{-1}$. Values obtained from iodine-128 are possibly less reliable since the short half-life of this isotope leads to a greater sensitivity to timing errors and to any time-dependent variations of flux during irradiation. A more important disadvantage is that iodine-127 has many very pronounced resonances in the energy range 10–2000 eV and the presence of incompletely moderated neutrons in our system makes the use of the thermal neutron activation cross-section rather inappropriate. The value of 6.4 barns is almost certainly too small to reflect the contribution made by neutrons of higher energy and thus leads to an overestimation of the neutron flux. In addition iodine-126 was observed in both heavy and light water solutions from the reaction $^{127}I(\gamma n)^{126}I$.

2. Irradiation of whole foods (Table 2)

No activity could be detected in irradiated codfish 1 h after the end of irradiation, although manganese-56 was observed in the monitor solutions located throughout the sample in discrete sealed vials. Manganese-56 was also produced in the case of irradiated rice and meat where the monitor was intimately mixed with the foodstuff, or contained in vials, although no activity was detected in the matrix. Clearly neutrons are generated in these systems. As expected, activity levels were higher in samples enriched with heavy water.

3. Irradiation of shielding and constructional materials

Examination of the activity produced in the manganese monitor indicated that the concrete did not generate a detectable flux of photoneutrons, and only traces of manganese-56 were induced by

Table 1. Irradiations of aqueous solutions

Experiment	Average dose (kGy)	Monitor isotope	Activity at the end of irradiation (Bq g⁻¹ monitor)	Neutron flux (n cm⁻² s⁻¹)	$[\text{n cm}^{-2}\text{ s}^{-1}][\mu\text{g(D) cm}^{-3}]^{-1}[\text{kGy s}^{-1}]^{-1}$	
10 MeV electrons						
D_2O(23 cm³)	20	I-126	2.2[b]	—	—	
NaI (0.1 M)		I-128	102.5[a]	6223	2.52	
$MnSO_4$(0.1 M)		Mn-56	10.05	551	0.225	
D_2O (23 cm³),	20	Na-24	< 40	—	—	
NaCl (0.1 M)		Cl-38	< 30	—	—	
0–10 MeV X-rays						
D_2O (23 cm³)	17	I-126	410[c]	—	—	
NaI (0.1 M)		I-128	59153[a]	3,577,337	1714	
$MnSO_4$(0.1 M)		Mn-56	13091[c]	717,779	344	
D_2O (23 cm³)	15.6	Na-24	200	634,000	324	325 (mean)
NaCl (0.1 M)		Cl-38	450	587,250	307	
0–10 MeV X-rays (control solutions)						
H_2O (60 cm³)	12.5	I-126	360[c]		—	
NaI (0.1 M)		Mn-56	185[a]	10,143	87,465	
$MnSO_4$ (0.1 M)						
H_2O (70 cm³)	13.8	Mn-56	207	11,350	88,647	104,888 (mean)
$MnSO_4$(0.2 M)						
H_2O (575 cm³)	9.0	Mn-56	211[a]	11,569	138,552	
$MnSO_4$(0.1 M)						

[a]Mean of two determinations.
[b]Mean of three determinations.
[c]Mean of four determinations.

Table 2. Irradiation of food samples

Food sample with manganese sulphate monitor (0.1 M)	Average dose (kGy)	Activity at the end of irradiation (Bq g^{-1} monitor)	Neutron flux (n cm^{-2} s^{-1})	[n cm^{-2} s^{-1}][μg(D) cm^{-3}]$^{-1}$ [kGy s^{-1}]$^{-1}$	
Rice (150 g)					
(i) H_2O (50 cm^3) D_2O (50 cm^3)	14	68,750	3,769,562	10,963	
(ii) H_2O (100 cm^3)	14	168	9,211	96,838	85,100 (mean)
Rice (166.5 g) H_2O (125 cm^3)	8.8	80	4,386	73,362	
Meat (745 g)					
(i) H_2O (35 cm^3; monitor solution in (7 vials)	11.7	417	22,864	210,632	179,517 (mean)
(ii) H_2O (60 cm^3)	13.5	339	18,587	148,402	
(iii) D_2O (60 cm^3)	13.5	73,370	4,022,877	32,508	
Fish (198 g) H_2O (20 cm^3; monitor in 4 vials)	8.8	87	4770	58,427	

irradiation of the brass and steel targets. Much higher monitor activities ($\times 30$) were observed when lead was tested. From a practical point of view it would appear that the only common shielding material which should be avoided in the vicinity of the electron beam is lead. The high atomic number leads to rather efficient conversion and all the stable isotopes of lead have (γn) thresholds well within the range of bremsstrahlung energies generated by 10 MeV electrons;

[^{204}Pb, 1.5%, 8.4 MeV; ^{206}Pb, 23.6%, 8.1 MeV;

^{207}Pb, 22.6%, 6.7 MeV; ^{208}Pb, 52.5%, 7.4 MeV].

In steel only iron-57 (2.2%, 7.6 MeV) has a threshold less than 10 MeV, accounting for the small observed neutron signal. Similarly in brass only zinc-67 (approx. 2%, 7.1 MeV) will contribute significantly to neutron production. Copper-65, with a threshold of 9.9 MeV, will contribute little. For concrete, we may take the composition to be that of typical crustal rock material, in which case only three isotopes, silicon-29, magnesium-25 and iron-57 have thresholds less than 10 MeV, their combined contribution to the mass of concrete being only 1.6%.

DISCUSSION

Results obtained from the 10 MeV electron irradiation of solutions of sodium iodide/manganese sulphate, and sodium chloride in heavy water show clearly the possibility of low levels of both photoneutron and neutron capture radioactive products, viz. iodine-126, iodine-128 and manganese-56, but no neutron activation product could be observed in light water solutions irradiated with electrons to a dose of 20 kGy.

The low level of activity observed is presumably mainly due to the poor efficiency of conversion to bremsstrahlung in the low atomic number target. Conversion in heavy and light water is $<4\%$ at 10 MeV and $<1\%$ at 2 MeV, fairly close to that in meat (taken as adipose tissue). This limits the contribution of both direct and indirect activation.

Adventitious X-ray can also arise from the electron beam in a variety of ways, e.g. collision of the beam with external constructional or shielding matter, or with the internal surfaces of the drift tube as a result of beam misalignment. Two general undesired effects may result; firstly, if the accidental target is of high atomic number then there may be substantial conversion to high energy bremsstrahlung, and secondly, the target material may contain isotopes of low photoneutron thresholds and a neutron flux may result. Both these criteria are met by the common shielding material lead and so the only neutron flux problem is likely to occur from the use of lead shielding in such a position where it may encounter the electron beam.

We may use the data in Table 1 to calculate the degree of direct (γn) activation of iodine, assuming an average concentration of that element in food (beef) of 3.5×10^{-2} ppm (Koch and Eisenhower, 1967). At this concentration the specific activity at the end of a 10 kGy irradiation would be less than 3.85×10^{-5} Bq kg^{-1}. The degree of direct activation depends upon the (γn) threshold but in this case the low activity is more a reflection of the low concentration of iodine in food. The indirect (γn) iodine-128 activity detected is 1.35×10^{-7} Bq kg^{-1} and for manganese-56 a value of 7.6×10^{-8} Bq kg^{-1} is generated by the 0.2 ppm concentration of manganese present in beef (Becker, 1983). Much higher levels of sodium and chlorine are present in foodstuffs (respectively 750 and 560 ppm) but even at these concentrations our Cerenkov measurements of the (nγ) activities (made after irradiation in the presence of very large amounts of deuterium which would maximise the effect of any adventitious photons) still predict that specific activities at the end of the irradiation cannot be distinguished from background. Even if the extreme

and highly unlikely view is taken that all the observed Cerenkov signal is due to sodium-24 or chlorine-38, and no allowance is made for background, the induced level of activity would not exceed 15 and 8.4 Bq kg^{-1} in food.

In fact, the activities induced in the solutions by the 10 MeV beam were so low, even under the artifically high conditions of monitor and deuterium concentrations, that it is extremely unlikely that electron irradiation of food would lead to any significant induction of radioactivity, even immediately after the irradiation. Indeed this has been confirmed by experiments involving electron treatment of beef (Martin and Becker, 1976; Miller and Jensen, 1987), and also by the recent electron studies on pepper (Furuta *et al.*, 1990). However, once the energy of electrons exceeds 10 MeV the (γn) thresholds of several minor, and more important, some major isotopes is exceeded.

Much higher levels of activity than were observed in the case of electrons were induced by the bremsstrahlung beam generated by 10 MeV electrons in a copper target, approx. 245 times as much iodine-126 being produced in both light and heavy water solutions. This corresponds to a mean specific activity of 9.26×10^{-3} Bq kg^{-1} food. The upper energy used in these experiments allows induction of species (e.g. iodine-126) which would not be generated in a beam whose energies are lower than the recommended ceiling of 5 MeV, but clearly the 10 MeV electron beam can generate as much as 0.4% of iodine-126 as the bremsstrahlung beam.

Adherence to a 5 MeV energy limit for X-rays eliminates the possibility of direct (γn) activation of any isotope whose threshold energy is greater. Only four isotopes can be activated at energies below 5 MeV and all have stable products; however, the reactions generate neutrons which could lead to indirect activation. This indirect effect may reasonably be neglected where electron beam irradiation is involved but must be considered for X-rays, for which a neutron flux of 1.44×10^3 times greater is observed. Of the four possible target nuclides, beryllium is unlikely to be found in food but carbon-13, deuterium and oxygen-17 are always present. With a threshold of 4.95 MeV the photoneutron reaction in carbon-13 is not likely to be significant, both on account of the low cross-section near the threshold and also because of the low abundance of high energy photons in a bremsstrahlung spectrum whose upper limit is 5 MeV; the situation is similar for oxygen-17 with a threshold of 4.2 MeV.

In any event the neutrons from deuterium would still, potentially, present a problem since the threshold is only 2.2 MeV and since deuterium occurs to the extent of 0.015% in all hydrogen. Thus, at any photon energy in excess of 2.2 MeV, there will inevitably be some slight neutron induced activity in a real food. We have estimated the deuterium-derived neutron flux generated in rice to be approx. 1.1×10^4 [n cm^{-2} s^{-1}][μg(D) cm^{-3}]$^{-1}$[kGy s^{-1}]$^{-1}$ and in meat 3.25×10^4 [n cm^{-2} s^{-1}][μg(D) cm^{-3}]$^{-1}$ [kGy s^{-1}]$^{-1}$ when these foods are irradiated with 0–10 MeV bremsstrahlung. Using the cross section vs energy profile for this reaction (IAEA, 1974) it is unlikely that these values would be reduced by more than a factor of two if X-rays of energy within the recommended ceiling were to be used.

The values quoted above are those derived from samples which have very high deuterium content and where virtually all the neutrons originate from the deuterium. In cases where no heavy water was deliberately added the total neutron flux measured is, of course, much lower and measurements less accurate. However, when expressed as a flux per unit concentration of deuterium per unit dose rate a higher value (approx. 5.5 times for meat) is obtained. Clearly there is at least one source of photoneutrons other than the reaction D(γn)H. At the bremsstrahlung energies used in this study the source cannot be unambiguously identified. It could be any of those discussed above, including carbon-13, and may even be absent altogether at X-ray energies within a 5 MeV ceiling.

The data given in Table 2 allows us to calculate the neutron flux generated in a sample irradiated in a flux of photons of energy in the range 0–5 or 0–10 MeV simply from a knowledge of the deuterium content of the sample, but only if the general background flux of neutrons is neglected. In order to make a practical estimate of the likely consequences which errs on the side of safety it is advisable to use the largest neutron flux value for calculation of activities, i.e. 1.79×10^5[n cm^{-2} s^{-1}][μg(D) cm^{-3}]$^{-1}$[kGy s^{-1}]$^{-1}$. Rather than make detailed calculations for each real activable impurity likely to be present in foods, we may calculate the activity which would be induced in a notional isotope of varied half-life and elemental thermal neutron activation cross-section.

Consider 1 dm^3 of food material containing "P" ppm. of an impurity, able to undergo neutron activation by (nγ) to give a radioactive nuclide whose half-life is $t_{1/2}$, and which has a cross section for activation of σ barns.

The activity produced is:

$$\frac{0.6\,\mathrm{P}\sigma \times 10^{-3}\,\phi}{\text{At. wt}} \times \frac{0.693\,t_{\text{irr.}}}{t_{1/2}}\ \text{Bq dm}^{-3}.$$

If we assume an impurity atomic weight of approx. 42 this expression reduces to $10^{-5}\,\mathrm{P}\,(\sigma/t_{1/2})\Phi t_{\text{irr.}}$ For a dose of 10 kGy, and for an average deuterium content equivalent to that of water, (16.7 μg cm^{-3}) the value of $\Phi t_{\text{irr.}}$ is given by the flux values [n cm^{-2} s^{-1}][μg(D) cm^{-3}]$^{-1}$[kGy s^{-1}]$^{-1}$ of Table 2 multiplied by 167. Assuming that the foodstuff has a specific gravity of unity, the activity (Bq kg^{-1}) is $167 \times 10^{-5}\,\mathrm{P}(\sigma/t_{1/2}) \times$ (neutron flux per unit deuterium concentration per unit dose rate).

We may consider a hypothetical nuclide whose half-life varies between 1 h and 10 days and whose neutron activation cross section varies between 1 and 100 barns, a range which covers all probable values. We have not concerned ourselves with nuclides whose half lives are shorter than 1 h because we expect the greater end-of-irradiation activity would be more than compensated by the rapid decay in any realistic time period needed for transportation and sale after irradiation: half-lives longer than 10 days preclude any serious activation. Choosing the value of $1.79 \times 10^5 [\text{n cm}^{-2}\,\text{s}^{-1}][\mu\text{g(D) cm}^{-3}]^{-1}[\text{kGy s}^{-1}]^{-1}$ for meat we obtain the end-of-irradiation activity values shown in Table 3.

The number of impurities and the exact value of "P" will depend on several factors but a rough estimate of induced activity may be obtained by assuming ten elements each present to the extent of 10 ppm. and having the cross section of half-life characteristics assumed above. The specific activities given in Table 3 should therefore be 100 times greater. Taking neutron leakage into account we may further increase the thermal neutron yield in our experimental food samples by a factor of 5 to approximate that in a bulk food.

Our upper estimates for the end-of-irradiation activities based on a cross-section of 100 barns and on half-lives of 1 h, 1 day and 10 days now become 4,165,173, and 17.3 Bq kg^{-1} respectively. The activity resulting from the assumption of a 1 h half-life would decrease to the 1 day value in only 4.6 h. These specific activities are upper limits and do not entirely result from deuterium-generated neutrons. Calculations based entirely on deuterium-derived neutrons would give activity levels approx. 5.5 times lower. In a commerical irradiation the extraneous neutrons observed in our study may be absent, only neutrons from natural deuterium being produced. Nevertheless, in the commissioning of a commercial plant it would be prudent to use flux monitor techniques to assess the magnitude of any intrinsic or extrinsic fluxes. If the neutron flux is known and if it can be assumed that all the induced activity arises as a result of neutron activation, then any foodstuff which is a candidate for radiation processing could be 'screened' for the possibility of radioactivity by neutron irradiation at high-flux in a reactor, any activity induced during the irradiation being calculated pro-rata.

We may compare our end-of-irradiation value of 173 Bq kg^{-1} (based on products with a half-life of 1 day) with the activity due to naturally occurring radionuclides which form an inseparable and inescapable component of food, e.g. carbon-14 and potassium-40. The former has a specific activity of 0.22 Bq g^{-1} of element and a typical carbohydrate such as starch therefore has an activity of about 98 Bq kg^{-1}. Potassium-40 has a specific activity of approx. 3 Bq g^{-1} of element, equivalent to an average activity of 12 Bq kg^{-1} of food. It is therefore reasonable to assume a natural radioactivity level in the region of 100 Bq kg^{-1} food; this assumption is in agreement with published data (Urbain, 1986).

Table 3. End-of-irradiation specific activites (Bq kg^{-1}) per ppm of notional isotope

	Half-life		
Cross-section (barns)	1 h	1 day	10 days
1	8.33×10^{-2}	3.47×10^{-3}	3.47×10^{-4}
100	8.33	3.47×10^{-1}	3.47×10^{-2}

Table 4. Estimated radioactivity induced in beef irradiated with 0–10 MeV X-rays (10 kGy)

	Specific activity (Bq kg^{-1})		
(nγ) product	At end of irradiation	After 6 h	After 1 day
Sodium-24	3.98	3.02	1.13
Silicon-31	1.58×10^{-3}	3.23×10^{-4}	—
Phosphorus-32	1.20×10^{-1}	1.18×10^{-1}	1.11×10^{-1}
Sulphur-35	1.2×10^{-3}	1.2×10^{-3}	1.2×10^{-3}
Chlorine-38	9.14	1×10^{-2}	—
Potassium-42	2.76	1.97	7.2×10^{-1}
Manganese-56	6.5×10^{-2}	—	—
Copper-64	7.86×10^{-3}	5.66×10^{-3}	2.12×10^{-3}
Zinc-69	3.47×10^{-1}	4×10^{-3}	—
Selenium-81	4.26×10^{-3}	—	—
Bromine-80	1.62	—	—
Bromine-82	3.27×10^{-3}	2.9×10^{-3}	2.0×10^{-3}
Rubidium-88	3.63×10^{-2}	—	—
Total	18.09	5.13	2.14

On the above reasoning the end-of-irradiation activity induced in foods by neutron activation is of the same magnitude as the natural level due to carbon-14 and potassium-40. The effective dose equivalent resulting from ingestion of our hypothetical isotopes cannot be calculated without some reference to their metabolism, and to allow a safe margin for differences in radiotoxicity we may make a worst case assumption, viz. that the induced activity has an Annual Limit on Intake of 10^5 Bq, a value comparable with but lower than those for particularly toxic beta emitting nuclides which have appreciable retentions in the body *cf* strontium-90 (ALI = 10^6 Bq), iodine-131 (ALI = 10^6 Bq), and iodine-126 (ALI = 8×10^5 Bq). To achieve this intake would necessitate an annual consumption of 578 kg of irradiated food; such a consumption, while not impossible, is highly improbable. The concept of ALI is not, of course, strictly applicable to the exposure of the general population but the calculation serves to show the magnitude of the expected dose.

More specifically, it is of interest to compare the activities induced in the notional isotopes with those calculated for a real food. Combining the neutron flux measurements made in our study with the composition of beef given by Becker we obtain the activities, uncorrected for possible neutron leakage, shown in Table 4.

These activities are the principal ones expected from the major matrix elements and biologically significant trace elements present in beef. However, we may speculate further on the possible effects of

Table 5. Comparison of activities induced by 0–10 MeV X-rays (10 kGy)

	Activities (Bq kg^{-1})			
	At end of irradiation	6 h	1 day	10 days
Notional isotope†	[a]173	145	86.5	0.17
(σ = 100 barns)	[b]4165	65	—	—
Beef‡	90	25.6	10.7	0.35
NRPB estimates (ACINF, 1986)				
(a) using maximum credible isotopic concentrations			35	16
(b) using realistic isotopic concentrations			0.12	1.2×10^{-2}
NRPB estimates (House of Lords) 1989				
Meat	130§	31	11	
Fish	190§	34	12	
Cereals	130§	27	10	

†Assumptions as in text: [a]$t_{1/2}$ = 1 day; [b]$t_{1/2}$ = 1 h.
‡Calculated using measured neutron fluxes obtained in this study and assuming the isotopic composition of beef according to Becker (1983).
§5 min after irradiation.

trace elements not normally tested for since they have no biological role. Useful candidates to consider in this respect are the lanthanides and associated elements yttrium and lanthanum since they are widespread, occur together in fairly well-defined proportion, and many have high neutron activation cross-sections. If we assume (Birch *et al.*, 1986) that beef has an inorganic ash content of 1% and further assume that the rare earth composition of this fraction is similar to that of the crustal rock of the Earth, then the total rare earth activity at the end of a 10 kGy irradiation is approx. 8×10^{-2} Bg kg^{-1}. The main contributors are cerium-143, cerium-141, samarium-152, scandium-46, europium-152 m, europium-152, erbium-171, holmium-166, yttrium-175, yttrium-90 and lanthanum-140. Thus the radioactive content of beef is little enhanced by the assumed presence of the lanthanides. Allowing a factor of 5 for possible neutron leakage we obtain a potential activity of 90 Bq kg^{-1} beef at the end of irradiation. This end-of-irradiation activity is comparable to the level of natural radioactivity due to potassium-40 and carbon-14 but reduces to a small fraction after only 1 day.

It is of interest to compare (Table 5) these activities with the NRPB activities (ACINF, 1986) calculated (using 'maximum credible' isotopic composition and 'realistic' isotopic composition) for canned meat and vegetables irradiated to 10 kGy. Our induced activity calculated for the 'notional' isotopes is greater than the NRPB estimates after one day but is well within their upper limit at 10 days after irradiation. Using our flux values to calculate the activity induced in beef we obtain activities at both one day and 10 days after irradiation which lie between the extreme values given by NRPB.

More recently the NRPB have revised their data and have calculated "worst case" activity values in a range of foods at 5 min, and at 1,3,6 and 12 h, after irradiation with cobalt-60 γ-ray photons and electrons and X-rays of 5 and 10 MeV (House of Lords, 1989). Calculated activity levels due to 10 MeV X-ray irradiation of meat, fish and cereals are included in Table 5. These values are comparable with those obtained for beef using the neutron fluxes measured in this study. The notional isotope with assumed half-life of one day gives a similar end-of-irradiation activity but this decays more slowly than the activities induced in the foods.

CONCLUSIONS

The radioactivity induced in food by electrons of energy up to 10 MeV is trivial and may be disregarded. However, there is evidence that adventitious X-rays produced by the beam can produce activity and for this reason there should be no increase in the proposed ceiling of 10 MeV, since at only slightly higher energies some major elements present in food can enter into activity-producing photon-induced nuclear reactions. Where LINAC generators are used the limiting energy should be the peak pulse energy and not the nominal energy.

When 0–10 MeV bremsstrahlung is used (and there is the possibility of some direct photoneutron activation) the induced activity is extremely small. The end of irradiation activity is of the same order as the radioactivity occurring naturally in food but because of differences in the radiotoxicity of different isotopes simplistic comparisons should be treated with care. The considerable decay of activity occurring within 24 h suggests that a post-irradiation delay of this duration would be beneficial. The recommended 5 MeV ceiling on X-ray energy for commercial irradiations removes the possibility of direct photon activation although it could be argued that a slightly lower energy would have the additional advantage of eliminating neutron production by the reactions $^{13}C(\gamma n)^{12}C$ and $^{17}O(\gamma n)^{16}O$ which have thresholds at 4.95 and 4.2 MeV respectively.

Appl. Radiat. Isot. Vol. 43, No. 5, pp. 567–575, 1992
Int. J. Radiat. Appl. Instrum. Part A
Printed in Great Britain

0883-2889/92 $5.00 + 0.00
Pergamon Press plc

Experimental Electron Beam Irradiation of Food and the Induction of Radioactivity

D. J. S. FINDLAY, T. V. PARSONS and M. R. SENE

Harwell Laboratory, AEA Technology, Harwell, Oxfordshire OX11 0RA, England

(Received 4 September 1991; in revised form 21 October 1991)

Samples of chicken, prawns, cheeses and spices were irradiated on the Harwell electron linear accelerator HELIOS at 20 MeV to assess mechanisms for the induction of radioactivity. The induced radioactivity was measured using a lead shielded Ge(Li) γ-ray spectrometer, and the results were compared with activities calculated on the basis of photoneutron and photoproton reactions induced by real and virtual photons. In general, there was good agreement. Bounds were also placed on the induction of radioactivity by capture of neutrons produced in the food samples themselves. Further, the data were used to assess the effects of a gross malfunction of an electron beam irradiation facility; after 1 day, the specific activity of food samples irradiated to 10 kGy at 20 MeV was ~0.01 Bq g^{-1}. In addition, food samples were also irradiated at 10 MeV, and irradiated and control samples were analysed for microbiological burden. Reductions in the microbiological burden of the food samples by factors consistent with those found in previous measurements were found.

1. Introduction

Food irradiation is a well established process for preserving food. Substantial pioneering work was carried out ~25 years ago by the United Kingdom Atomic Energy Authority at its Wantage Research Laboratory (Sharpe, 1990). Most of the food irradiation plants in use around the world are radioisotope plants based on ^{60}Co. However, some electron beam irradiation facilities have recently been commissioned. Electron beam facilities are perceived to have advantages from the general public's point of view as they do not incorporate large permanent radioisotope inventories. When an electron accelerator is switched off, little or no residual radioactivity remains.

The main aim of carrying out the work described in this paper was to assess mechanisms for the induction of radioactivity in food by electron beam irradiation. For reasons of public acceptability of electron beam food irradiation, it is important that these mechanisms can be shown to be understood. Accordingly in this paper is described the measurement and calculation of radioactivity in food induced by electron beam irradiation. To produce measurable radioactivity, the irradiations were carried out at 20 MeV, since at the regulatory maximum energy at 10 MeV the induced radioactivity is essentially zero. *It must be emphasized that 20 MeV irradiations are completely different from practical food irradiation at up to 10 MeV and were only carried out so that induced radioactivities could be measured.*

Previous relevant work includes that of Miller and Jensen (1987), Becker (1983), Leboutet and Aucouturier (1985), Fleming (1985), Rogers and Whittaker (1970) and von Janetschke *et al.* (1985). However, the works of Becker and of Leboutet and Aucouturier are restricted to calculations, and those of Fleming and of Rogers and Whittaker to ^{60}Co irradiations. There does not appear to have been undertaken a comprehensive comparison of measured and calculated radioactivities in electron beam food irradiation. The work described in this paper is intended as a contribution towards such a comparison.

2. Experimental Arrangements

2.1. Electron beam characteristics

Electron beam irradiations were carried out at 10 and 20 MeV on the Low Energy (LE) beam line of the Harwell electron linear accelerator HELIOS (Lynn, 1980). The LE beam is an energy-analysed electron beam, obtained by bending the beam through 90° from the accelerator axis. The bend incorporates energy-defining slits, and these were set to ±5%. The energy calibration (Findlay *et al.*, 1986) of the beam line has been established absolutely by relating measured photoneutron thresholds to the magnetic field in the bending magnets measured using a radiation-resistant Rawson–Lush rotating-coil gaussmeter. By energizing a magnet ~5 m downstream from the 90° bend the electron beam was bent up at

an angle of 30° and emerged into air through a thin window. After the thin window a computer-driven scanning magnet was used to scan the electron beam in a raster pattern over a ~30 × 30 cm square area ~117 cm from the scanning magnet. During the irradiations, the accelerator was run at 25 pps, producing an energy-analysed beam of ~2.5 μs, ~250 mA pulses.

2.2. Samples and irradiations

Samples of chicken, prawns, cheeses and spices were obtained from local supermarkets. The chicken (both thighs and drumsticks) and the prawns were bought frozen, and held in a freezer until after irradiation. Two separate bags each of chicken thighs and prawns were bought, and one thigh and prawn from each bag irradiated together. The cheeses, boursin and brie, were frozen immediately after purchase. The spices chosen were pepper and turmeric.

Frozen samples were removed from a freezer immediately after irradiation, and were taped in thin, labelled polythene bags on a thin, ~30 × 30 cm, perspex sheet. Immediately after irradiation the samples were put back in the freezer. The irradiations varied in duration, but at 10 MeV the maximum time between removing the samples from the freezer before irradiation and returning the samples to the freezer after irradiation was 5 min, and the temperature of the samples did not rise above 0°C.

2.3. Dosimetry

To establish the degree of uniformity of the scanned beam, a 5 × 5 array of Faraday cups on 5 cm centres were constructed. The aluminium cups had front apertures 2.5 cm in dia, 1 cm thick walls and a base thickness of 2.5 cm. The cups were embedded in perspex, and were enclosed in a 1.6 mm thick steel box. The front of the box was a 2.5 cm thick aluminium collimator plate with 2.5 cm dia holes corresponding to the entrance apertures of the individual Faraday cups. The currents deposited in the 25 Faraday cups were taken to earth through parallel RC-combinations of 1 MΩ‖2.2 μF, and a high input impedance (500 MΩ) 200 mV f.s.d. digital voltmeter (DVM) was placed across each RC-combination in turn. Typical DVM readings were 50 mV. Considering the 5 × 5 array of cups as a central cup, an inner square of 8 cups and an outer square of 16, for the 10 MeV irradiations, the variation over the inner square was $^{+0}_{-8}$% and over the outer square was $^{+0}_{-27}$%.

Since the Faraday cup array has to be removed while food samples are irradiated, a secondary beam monitoring system was set up. This simple but effective system was a 1 VA^{-1} toroidal current transformer* placed immediately after the vacuum window through which the electron beam emerges into air but before the scanning magnet, an amplifier to raise the signal to ~10 V, and a current integrator fed by the amplifier via a low leakage silicon diode. Previous off-line tests in the laboratory had shown that the response of this system was independent of pulse repetition frequency to ±5%. This secondary system was calibrated in terms of the *mean* electron fluence established absolutely from the Faraday cup measurements.

Irradiation dose rates were computed from the measured electron fluxes assuming a $dE/d(\rho x)$ value of 2.00 MeV cm^2 g^{-1}. This value for the collison stopping power is accurate to 5% water, muscle and bone between 1 and 10 MeV, and its use implies the relationship 10 kGy ≡ 5 μC cm^{-2} as the equivalence of radiation dose and electron fluence.

During irradiation, Harwell Gammachrome YR perspex dosimeters (Whittaker, 1990) were positioned alongside the food samples. These were read after the irradiations had been carried out. The results at 10 and 20 MeV were on average (5 ± 10)% lower than the values expected from the Faraday cup calibrations.

It is clear that the dosimetry established for the work described in this paper is only accurate to ~20%. However, such an accuracy is satisfactory since, as stated below, the calculations of induced radioactivity with which the data are compared are themselves only accurate to ~30%. *In particular, the results given in this paper should not be regarded as a calibration of the Harwell Gammachrome YR perspex dosimeters.*

At the beginning of the 20 MeV irradiations, which were carried out after the 10 MeV irradiations, the scanning magnet developed an electrical fault. Consequently, a 1 mm thick aluminium scatter plate positioned 9.5 cm before the scanning magnet was used instead to produce a large area beam. The Faraday cup array was used again to measure the electron fluxes on the samples, although at 20 MeV neither the collimator plate nor the base of the cups are sufficiently thick to stop the electrons completely. Only an area bounded by the inner square of 8 cups (~10 × 10 cm) was used at 20 MeV, and the variation over the inner square was $^{+0}_{-32}$%.

2.4. Counting system

The food samples were counted using an 80 cm^3 Ge(Li) γ-ray spectrometer surrounded on all sides by 10 cm of lead shielding. The efficiency of the system as a function of distance from the Ge(Li) crystal had been calibrated using standard sources including a ^{152}Eu source. The food samples irradiated at 20 MeV occupied roughly spherical volumes of 320 ± 40 cm^3 and were placed against the front surface of the spectrometer. The efficiency for counting the samples was obtained by integrating over the volume occupied by the samples and included the effects of γ-ray absorption in the sample. At 1 MeV the efficiency was 2.27 ± 0.45 × 10^{-3} counts per γ-ray emitted from the sample, and varied with γ-ray energy roughly as $E^{-0.94}$. The large error on the efficiency is to accommodate uncertainties in the precise configuration of the food being counted.

*100 turns on a 0.05 mm mu-metal tape toroid fed into a 100 Ω coaxial cable.

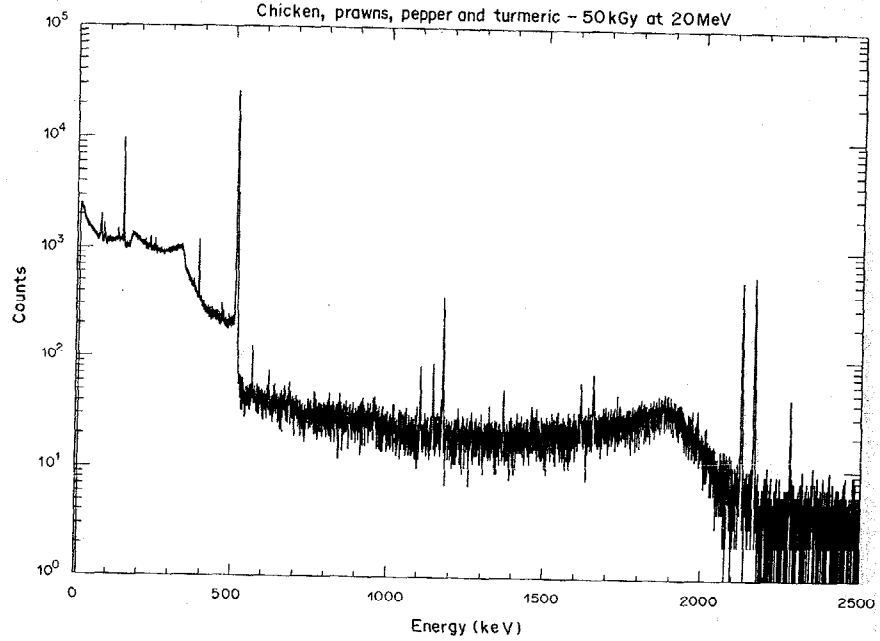

Fig. 1. The Ge(Li) γ-ray spectrum corresponding to the first of the 5000 s counting periods listed in Table 1.

The Ge(Li) γ-ray spectrum corresponding to the first of the 5000 s counting periods (see Section 3.1) for chicken, prawns, pepper and turmeric is shown as Fig. 1.

3. Analysis

3.1. Sample counting

Samples irradiated at 20 MeV were counted using the 80 cm^3 Ge(Li) γ-ray spectrometer described in Section 2.4. The timetable of the irradiation and counting procedures is summarized in Table 1. The measured γ-line energies and half-lives are given in Table 2 for the chicken, prawns, pepper and turmeric samples. From the supplementary experiment described in Section 4.1, it is clear that the strontium is in the prawns. Absolute values for the specific activity are also given. Similar quantities for brie and boursin are given in Table 3. It should be noted that the consistency of the results for different γ-lines from the same nuclide gives confidence that the extended source representation of the food samples was correct.

Table 1. Timetable of irradiation and counting of 20 MeV food samples

Irradiated on 12 Sept. 1990	Samples counted	Counting sequence
1713–1720 h	One 120 g drumstick and two 30 g prawns	Counted together; 3 × 5000 s sequence, starting at 1810
1730–1737 h	15 g pepper and 15 g turmeric	
1743–1750 h	98 g brie and 72 g boursin	Counted together; 5 × 5000 s sequence starting at 2320

The ^{13}N activity was obtained from the 511 keV annihilation-γ line in the first of the Ge(Li) spectra summarized in Table 1 after subtraction of the ^{11}C, ^{34m}Cl and ^{38}K 511 keV components. The subtrahends were obtained from the measured γ-ray lines for ^{34m}Cl and ^{38}K and from calculations (see Section 4.1) for ^{11}C. [In the GE(Li) spectra for the cheese samples, the ^{11}C, ^{13}N and ^{38}K activities have all died away; this explains the equality of the 146 keV and 511 keV lines in Table 3.]

3.2. Calculation of induced activity

The primary sources of induced radioactivity in samples irradiated in electron beams at the energies of interest in the present work are photon- and electron-induced nucleon 'knock-out' reactions. For the configuration used in the present work the relevant mechanisms are:

—Photonuclear reactions induced by bremsstrahlung (real) photons produced by the passage of the electron beam through the aluminium scatter plate upstream of the samples.

Table 2. Measured γ-lines, half-lives, identifications, reaction thresholds and specific activities at 1737 h (immediately after irradiation of the pepper and turmeric) for chicken, prawns, pepper and turmeric irradiated to 50 kGy at 20 MeV

γ-line (keV)	Measured half-life (min)	Nuclide	Half-life	Reaction	S_n or S_p (MeV)	Specific activity (Bq g^{-1})
146	32 ± 1	^{34m}Cl	32.2 min	^{35}Cl(γ, n)	12.6	33 ± 7
231	92 ± 23	^{85m}Sr	68.0 min	^{86}Sr(γ, n)	11.5	0.36 ± 0.10
248	s	^{84m}Rb	20.5 min	^{85}Rn(γ, n)	10.5	1.7 ± 0.5
373	l	^{43}K	22.3 h	^{44}Ca(γ, p)	12.2	0.038 ± 0.010
388	164 ± 9	^{87m}Sr	2.81 h	^{88}Sr(γ, n)	11.1	1.27 ± 0.26
464	s	^{84m}Rb	20.5 min	^{85}Rb(γ, n)	10.5	1.13 ± 0.69
[a]511		^{13}N	10.0 min	^{14}N(γ, n)	10.6	510 ± 280
618	l	^{42}K	22.3 h	^{44}Ca(γ, p)	12.2	0.048 ± 0.012
1176	29 ± 1	^{34m}Cl	32.2 min	^{35}Cl(γ, n)	12.6	36 ± 8
1368	l	^{24}Na	15.0 h	^{25}Mg(γ, p) or ^{23}Na(n, γ)	12.1	0.13 ± 0.03
2128	32 ± 1	^{34m}Cl	32.2 min	^{35}Cl(γ, n)	12.6	37 ± 8
2168	s	^{38}K	7.6 min	^{39}K(γ, n)	13.1	680 ± 180

s, half-life short compared to 5000 s counting periods.
l, half-life long compared to 5000 s counting periods.
[a]After subtraction of ^{11}C, ^{34m}Cl and ^{38}K as described in Section 3.1.

Table 3. Measured γ-lines, identifications, reaction thresholds and specific activities at 1750 h for brie and boursin irradiated to 50 kGy at 20 MeV

γ-line (keV)	Nuclide	Half-life	Reaction	S_n or S_p (MeV)	Specific activity (Bq g^{-1})
146	^{34m}Cl	32.2 min	^{35}Cl(γ, n)	12.6	93 ± 25
373	^{43}K	22.3 h	^{44}Ca(γ, p)	12.2	0.010 ± 0.005
388	^{87m}Sr	2.81 h	^{88}Sr(γ, n)	11.1	0.033 ± 0.015
511	^{34m}Cl	32.2 min	^{35}Cl(γ, n)	12.6	99 ± 23
618	^{43}K	22.3 h	^{44}Ca(γ, p)	12.2	0.013 ± 0.005
1368	^{24}Na	15.0 h	^{25}Mg(γ, p) or ^{23}Na(n, γ)	12.1	0.09 ± 0.02

—Photonuclear reactions induced by bremsstrahlung (real) photons produced in the samples themselves by the passage of the electron beam through the samples.
—Electronuclear reactions induced by the electron beam as it passes through the samples.

The radioactivities induced in these ways are distributed differently throughout the samples. The real photons produced by the scatter plate interact throughout the whole thickness of the samples. Real photons produced in the samples themselves only interact at thicknesses greater than the sample thickness at which they were produced. Electron-induced reactions only occur towards the front of the samples where the electron beam energy is above the photonuclear thresholds at ~10 MeV.

The yields of nuclides in the food samples were calculated using the expressions in the appendix for y_j^γ, y_j^e and y_j^ν, respectively the number of atoms of nuclide j per cm^3 per electron produced by bremsstrahlung from the scatter plate, the number of atoms of nuclide j per electron produced by bremsstrahlung in the samples themselves, and the number of atoms of element j per electron produced from electron-nuclear interactions in the samples. The number of Y_j of atoms of nuclide j produced in the samples irradiated is obtained by multiplying y_j^γ by the volume of the sample and the number of electrons incident on the scatter plate (expressed in terms of the electron fluence at the sample and a geometrical factor), multiplying y_j^e and y_j^ν by the area of the sample and the electron fluence, and adding, according to

$$Y_j = \pi\overline{\theta_s^2}d^2\frac{q}{e}(m/\rho)y_j^\gamma + \frac{q}{e}\frac{(m/\rho)}{t}y_j^e + \frac{q}{e}\frac{(m/\rho)}{t}y_j^\nu$$

where $\sqrt{\overline{\theta_s^2}} = 0.075$ rad is the root mean square multiple scattering angle for the 1 mm aluminium scatter plate, $d = 126.5$ cm (Section 2), $q = 25\,\mu$C cm^{-2} for 50 kGy dose (see Section 2.3), $t = 2.5$ cm is the thickness of the samples, ρ is density of the samples, and m is the mass of the food samples, 210 and 170 g respectively for the chicken, prawns, pepper and turmeric (CPPT) sample and the brie and boursin (BB) sample. The data used for the cross-sections $\sigma_j(k)$ are given in Table 4, and, where relevant, a total cross-section was assumed to be split equally between cross-sections to the ground state and to the isomer. To accommodate the effects of the approximations used and uncertainties in the precise configurations of the food samples during irradiation, an error of 30%, derived from the effects of making realistic changes to the relevant parameters, was assigned.

Table 4. Details of the cross-section of $\sigma_j(k)$ used in the calculations described in Section 3.2

Reaction	Ref.	Notes
^{2}H(γ, n)^{1}H	(Bülow and Forkman, 1974)	
^{12}C(γ, n)^{11}C	(Berman, 1974)	
^{14}N(γ, n)^{13}N	(King *et al.*, 1960)	Excited states of ^{13}N p-unstable
^{16}O(γ, n)^{15}O	(Berman, 1974)	
^{25}Mg(γ, p)^{24}Na	(Forkman and Petersson, 1987)	Assumed $\sigma = 4\pi(d\sigma/d\Omega)_{\gamma p0+1}(90°)$ for ^{24}Mg
^{31}P(γ, n)^{30}P	(Bülow and Forkman, 1974)	
^{35}Cl(γ, n)^{34}Cl	(Forkman and Petersson, 1987)	
^{39}K(γ, n)^{38}K	(Bülow and Forkman, 1974)	
^{40}Ca(γ, n)^{39}Ca	(Forkman and Petersson, 1987)	
^{44}Ca(γ, p)^{43}K	(Oikawa and Shoda, 1977)	
^{85}Rb(γ, n)^{84}Rb	(Forkman and Petersson, 1987)	Assumed ^{nat}Rb
^{86}Sr(γ, n)^{85}Sr	(Forkman and Petersson, 1987)	Assumed ^{nat}Sr
^{88}Sr(γ, n)^{87}Sr	(Forkman and Petersson, 1987)	Assumed ^{nat}Sr

Table 5. Measured potassium content of food samples in units of percent by weight

Food sample	Potassium content
Chicken drumstick + 2 prawns	0.37 ± 0.06%
Pepper	1.56 ± 0.15%
Turmeric	3.11 ± 0.19%

In the present work, except for potassium, the elemental concentrations f_j of parents of nuclide j in the food samples were not measured. Instead the elemental concentrations assumed were taken from Lide (1990) when values were specifically given for the particular foodstuffs, and from the human body concentrations given in Anderson (1989) when no specific values were available. For the BB sample, the chlorine atomic concentration was assumed to be the same as the sodium concentration. The potassium concentrations were measured by counting ^{40}K using the Ge(Li) γ-ray spectrometer described in Section 2.4. The results are given in Table 5. The potassium content was obtained by comparing the ^{40}K 1461 keV peak in the Ge(Li) spectrum from the samples with the ^{40}K peak from appropriate samples of di-potassium hydrogen phosphate K_2HPO_4.

In Tables 6 and 7 are given comparisons of the measured and calculated activities. The distribution of strength amongst the three routes given above for the induction of radioactivity is roughly 1:2:1 (in the order that the three routes appear above).

4. Discussion

4.1. 20 MeV irradiations

From Tables 6 and 7, it can be seen that on the whole the measured and calculated specific activities are in good agreement. For the CPPT sample, the results for nitrogen, chlorine, potassium and calcium agree within the errors. For the BB sample the results for calcium agree well, but the calculated chlorine value is ~3 times greater than the measured value. As stated in Section 3.2 the chlorine concentration was taken from the sodium concentration, but it is clear that the processing steps in the production of cheese could alter the relative concentrations. For rubidium the measured result for the CPPT sample was ~3 times greater than the calculated result. For strontium, the measured results were ~10 times greater than the calculated results for the CPPT sample but ~3.5 times smaller for the BB sample. The CPPT strontium results are consistent with the ability of shellfish to concentrate such elements.

Although the concentrations of elements assumed in the present work are very similar to the concentrations measured for beef (Miller and Jensen, 1987), nevertheless it is clear that it would have been preferable for a full elemental analysis of each sample to have been carried out. However, except for the prawns which have already been considered and the sodium activity which will be discussed below, the results show that the measured induced radioactivities for all elements agree to within a factor of 3 with calculated values when elemental concentrations are taken from standard tables. This is the first time that such a comparison between measured and calculated activities has been made, and it is hoped that the present work should therefore prove useful in the context of expanding the use of electron beam irradiation of food.

In previous work (Miller and Jensen, 1987) the ^{24}Na activity induced in food by electron beam irradiation was shown to be much greater than expected, but the mechanism was not resolved, although neutron

Table 6. Comparison of measured and calculated values of specific activity for the CPPT sample

Element	Concentration assumed (% by weight)	Product nucleus	Specific activity (Bq g^{-1}) Measured	Calculated
N	2.6	^{13}N	510 ± 280	280 ± 80
Mg	0.027	^{24}Na	0.13 ± 0.03	0.014 ± 0.004
Cl	0.12	^{34m}Cl	36 ± 8	23 ± 7
K	[a]	^{38}K	680 ± 180	740 ± 220
Ca	[b]	^{43}K	0.043 ± 0.011	0.040 ± 0.012
Rb	0.00046	^{84m}Rb	1.4 ± 0.6	0.47 ± 0.14
Sr	0.00046	^{85m}Sr	0.36 ± 0.10	0.029 ± 0.009
		^{87m}Sr	1.27 ± 0.26	0.108 ± 0.032

[a]Fractions by weight from Table 5.
[b]Fractions by weight: drumstick + prawns, 1.4%; pepper, 0.36%; turmeric, 0.40%.

Table 7. Comparison of measured and calculated values of specific activity for the BB sample

Element	Concentration assumed (% by weight)	Product nucleus	Specific activity (Bq g^{-1}) Measured	Calculated
Mg	0.027	^{24}Na	0.09 ± 0.02	0.014 ± 0.004
Cl	1.23	^{34m}Cl	96 ± 24	320 ± 100
Ca	0.529	^{43}K	0.011 ± 0.005	0.017 ± 0.005
Sr	0.00046	^{87m}Sr	0.033 ± 0.015	0.115 ± 0.035

Table 8. Calculated neutron production within the CPPT sample for a 50 kGy dose at 20 MeV

Element	% by weight	Neutrons produced
H	10	5.16×10^7
C	23	1.23×10^7
N	2.6	4.89×10^8
O	61	7.89×10^8
P	1.1	2.03×10^8
Ca	1.4	3.51×10^7
		1.58×10^9

Table 9. Comparison of parameters for production of ^{42}K and ^{24}Na from naturally occurring ^{41}K and ^{23}Na respectively

Parameter	^{42}K	^{24}Na
Half-life $t_{1/2}$	12.36 h	14.96 h
γ-Line (keV)	1525	1368
γ-Line abundance (%)	18.4	100
Parent isotopic abundance (%)	6.73	100
Neutron capture resonance integral I_γ (b)	1.42 ± 0.06	0.31 ± 0.01
Parent concentration assumed (% by weight)	0.65[a]	0.14

[a]Appropriately weighted mean of values from Table 5.

production from iron and aluminium in and around the accelerator was postulated. In the present work, it can be seen from Tables 6 and 7 that the ^{24}Na activity is a factor of ~ 8 greater than that calculated from the $^{25}Mg(\gamma, p)$ reaction. [It is interesting to note that the ratio of the ^{24}Na and the ^{13}N activities in the present work and in Miller and Jensen (1987) are very similar.] However, ^{24}Na can also be produced by the neutron capture reaction $^{23}Na(n, \gamma)$ on the sodium occurring naturally as an important constituent of living tissue as well as by the $^{25}Mg(\gamma, p)$ reaction, and neutrons can be produced at electron energies above 2.2 MeV from the $^2H(\gamma, n)$ reaction on the deuterium naturally present in living tissue and at higher electron energies but still below 10 MeV from ^{13}C and $^{17,18}O$.

The results of a calculation of the total neutron production in the food samples is summarized in Table 8. These results were obtained using the human body elemental composition (Anderson, 1989) in the same way as the results tabulated in Section 3.2. Neutron production from all elements whose concentrations were greater than 1% were considered. Monte Carlo neutron transport calculations were then carried out using the computer program MORSE (Emmett, 1975). The food samples were represented as a cylinder 2.5 cm thick, 200 cm^3 in volume, and with a density of 1 g cm^{-3} made up of hydrogen, carbon and oxygen in the proportions (after re-normalization to 100%) given in Table 8, and the sodium concentration was assumed to be 0.14% (Anderson, 1989) by weight. Neutrons were assumed to be produced uniformly throughout the samples with an energy of 1 MeV. The result was $7.7 \pm 0.8 \times 10^{-7}$ ^{24}Na atoms produced per source neutron, leading to a specific activity of $7.5 \pm 0.8 \times 10^{-5}$ Bq g^{-1}. This is so much less than the measured value of 0.13 ± 0.03 in Table 2 that neutron production in the food sample itself followed by neutron capture on sodium in the food cannot be considered as a significant ^{24}Na production route*.

*It should be noted, however, that in general the fraction of neutrons captured depends on the sample size, the fraction increasing as the sample size increases. Such sample size dependence may explain the difference between the value $\sim 10^{-7}$ for the ratio of the activity of the ^{24}Na produced by neutron capture and the ^{13}N activity in the present work and the value $\sim 10^{-6}$ for Becker's results quoted by Miller and Jensen (1987).

Further evidence that some other production route dominates, and that the existence of a strong neutron source other than the production of neutrons in the food sample itself is excluded, can be obtained by looking for evidence of ^{42}K in the Ge(Li) γ-ray spectrum. If neutron capture produces ^{24}Na from naturally occurring sodium, then ^{42}K must also be produced from naturally occurring potassium. The relevant parameters for ^{42}K and ^{24}Na are given in Table 9. The neutron spectrum in the food sample is very under-moderated, and so it is appropriate to use the neutron capture resonance integral I_γ rather than the thermal capture cross-section σ_γ as a measure of the neutron capture probability. On the basis of Table 9, the calculated counts in the Ge(Li) spectrum for the 1525 keV ^{42}K γ-line should be 41 ± 4 for the observed 1368 keV ^{24}Na γ-line peak area of 194 ± 17. The counts actually observed at the position of the ^{42}K peak were 3 ± 3. This comparison shows that to explain the observed ^{24}Na activity on the basis of neutron capture would require the sodium concentration in the food to be *at least* several percent. Such a high sodium concentration is unrealistic. It is concluded therefore that the ^{24}Na activity must be due to the $^{25}Mg(\gamma, p)$ reaction, and therefore that either the magnesium concentration was substantially higher than the value of 0.027% assumed or a value which was too low was assumed for the $^{25}Mg(\gamma, p)$ cross-section.

To confirm this conclusion, a supplementary experiment was carried out. A 202 g sample of chicken (94 g), prawns (65 g), NaCl (25 g domestic salt), $Mg(OH)_2$ (5 g) and K_2HPO_4 (13 g) was irradiated to 200 kGy at 20 MeV on the axis of the Harwell electron linear accelerator HELIOS. (The chicken and prawns were from the same batches used in the original experiment.) The 9.8 g sodium content of the NaCl component was therefore 33 times greater than the sodium concentration in the original CPPT sample on the basis of the assumed 0.14% concentration. However, the ^{24}Na activity re-normalized to correspond to a 50 kGy dose obtained by counting the NaCl component alone after irradiation was very similar to the ^{24}Na activity in Table 2, and on the basis of the results of counting the $Mg(OH)_2$ sample was consistent with induction through the $^{25}Mg(\gamma, p)$ reaction on the known magnesium content of the salt (magnesium carbonate was present in the salt at the $\sim 1\%$ level to improve the free-running properties of the salt). Because the ^{24}Na activity did not scale with the sodium concentration, it is therefore clear that the ^{24}Na activity is not due to neutron capture on sodium in the food but to the $^{25}Mg(\gamma, p)$ reaction, which is contrary to the conclusions of Miller and Jensen (1987). On the basis of the ^{24}Na activity in the $Mg(OH)_2$ sample, the magnesium concentration of the chicken and prawn components was $0.075 \pm 0.025\%$ by weight. This is ~ 3 times greater than the concentration assumed in Tables 6 and 7, and so the $^{25}Mg(\gamma, p)$ ^{24}Na cross-section used in the calculations must have been too low by a factor ~ 2.6.

Table 10. Relevant parameters for neutron capture on elements with concentrations greater than 0.1%. Isotopes with abundances less than 0.1% and half-lives less than 1 h or greater than 1 year can be neglected

Element	% by weight	Abundance Isotope	(%)	Product	Half-life	I_γ (b)
O	61	^{18}O	0.2	^{19}O	27 s	
C	23	^{13}C	1.1	^{14}C	[a]5700 y	
H	10	^2H	0.015	^3H	[a]12.3 y	
N	2.6	^{15}N	0.37	^{16}N	7.13 s	
Ca	1.4	^{40}Ca	97	^{41}Ca	1.03×10^5 y	
		^{44}Ca	2.1	^{45}Ca	163 d	0.56 ± 0.01
		^{46}Ca	0.004			
		^{48}Ca	0.19	^{49}Ca	8.72 m	
P	1.1	^{31}P	100	^{32}P	[a]14.3 d	0.085 ± 0.010
S	0.2	^{34}S	4.2	^{35}S	[a]87.5 d	0.0015
		^{36}S	0.02			
K	0.2	^{41}K	6.7	^{42}K	12.4 h	1.42 ± 0.06
Na	0.14	^{23}Na	100	^{24}Na	15.0 h	0.31 ± 0.01
Cl	0.12	^{35}Cl	76	^{36}Cl	3.0×10^5 y	
		^{37}Cl	24	^{38}Cl	37.2 m	

[a]No γs.

As explained in Section 1, the main purpose of the present work was to confirm that the mechanisms for the induction of radioactivity in food were understood. All the nuclides identified in Tables 2 and 3 are produced by the ejection of a neutron (^{13}N, ^{34m}Cl, ^{38}K, ^{84m}Rb and $^{85m,87m}Sr$) or a proton [^{24}Na via the $^{25}Mg(\gamma, p)$ route, ^{43}K]. Such reactions have definite thresholds, and the thresholds are also given in Tables 2 and 3. It can be seen that in no case is the relevant reaction threshold below 10 MeV, and so none of these reactions can proceed for 10 MeV electron beam irradiation. The production of ^{24}Na is possible at 10 MeV via the $^{24}Na(n, \gamma)$ route because of neutron production from the $^2H(\gamma, n)$, $^{13}C(\gamma, n)$ and, to a lesser extent, $^{17,18}O(\gamma, n)$ reactions. The neutron production from ^2H was calculated for an electron-only flux (i.e. bremsstrahlung production in a scatter plate was excluded) assuming hydrogen (and consequently deuterium) concentration by weight of 10 and 0.003% respectively in the manner described in Section 3.2. Neutron production from ^{13}C and $^{17,18}O$ was neglected because of the dominance of neutron production from ^2H shown by Allen and Chaudhri (1988). For a 50 kGy dose, the number of neutrons produced was $2.0 \pm 0.6 \times 10^7$, which, from scaling the results of the MORSE calculation above, corresponds to a specific activity of $\sim 1 \times 10^{-6}$ Bq g^{-1}.

The effects of neutron capture on elements other than sodium may be estimated from Table 10 in which are tabulated relevant details of elements present with concentrations down to 0.1%. From an examination of Table 10 it can be seen that no routes more significant than the sodium route are expected.

The irradiations above the regulatory 10 MeV limit can be used to assess the effects of a gross malfunction of an electron beam irradiation plant. For economic reasons, an electron linear accelerator in an electron beam food irradiation facility has to operate at or near its maximum current. However, if the energy of the accelerator in this fully loaded condition is 10 MeV, the energy for light loading can approach 20 MeV. Therefore if a fault led to a substantial reduction of injected current, if the energy analysis system continued to transmit the electron beam, and if the current integration arrangements continued to operate, then a given radiation dose apparently delivered at "10 MeV" from a food irradiation accelerator could in fact be delivered at an energy approach 20 MeV. In Table 11 are shown activities 1 h and 1 day after irradiation to the regulatory limit of 10 kGy at 20 MeV derived from the measured activities tabulated in Tables 2 and 3. It can be seen that after 1 day the activity is only ~ 0.01 Bq g^{-1}. At this time, the activity is dominated by ^{43}K and ^{24}Na with ~ 1 day half-lives and so the contributions from these isotopes will continue to halve every ~ 1 day. Activities are not given in Table 11 at times greater than 1 day because such activities could be dominated by weak long-lived isotopes not detected in the present measurements. An activity of 0.01 Bq g^{-1} may be put into context by

Table 11. Specific activities of food samples after 1 day 1 h

Food sample	Specific activities (Bq g^{-1}) after 1 h	1 day
Chicken, prawns, pepper, turmeric	0.70	0.0135
Brie, Boursin	*	0.0068

*No measurements were made at short times.

noting that low-sodium salts for domestic use have natural specific activities of ~ 10 Bq g^{-1} (because of the potassium content), pepper and turmeric have specific activities of ~ 0.5 Bq g^{-1} (again because of the potassium content), and the limits imposed on lamb meat for consumption in the U.K. after the Chernobyl accident were also ~ 0.5 Bq g^{-1}.

4.2. 10 MeV irradiations

The food samples irradiated at 10 MeV were analysed for microbiological burden by the Leatherhead Food Research Association*, and the results may be summarized as follows. For chicken irradiated to a dose of 7 kGy, reductions in bacterial numbers of 2–3 orders of magnitude were found. For prawns irradiated to 3 kGy, the reductions were ~ 1.5 orders of magnitude. For boursin cheese irradiated to 2 kGy, no reduction was found, whereas for brie irradiated to 2 kGy 3 orders of magnitude reduction was found. For pepper and turmeric irradiated to 10 kGy, 4–5 orders of magnitude reductions were found. It can therefore be seen the mean value of the factors by which the bacterial numbers were reduced by irradiation was ~ 3 orders of magnitude. This is consistent with the values typically achieved in food irradiation: see Mossel (1985). The variability of the results achieved in the present work is most likely to be due to the small number of samples analysed (e.g. only one irradiated and one non-irradiated sample of each cheese type was analysed). It must be remembered that reductions in bacterial numbers were obtained by comparing each irradiated sample with a different non-irradiated sample. To achieve better results, a much larger number of samples would have to be analysed to ensure statistical convergence.

The only counting carried out of food samples irradiated at 10 MeV was a relatively short 1000 s count of a 340 g composite sample of two prawns, one chicken drumstick and one chicken thigh carried out within 1 h of irradiation. The limit of specific activity deduced from the absence of any peaks above background was ~ 0.01 Bq g^{-1} (a $\sim 1-\sigma$ limit). This is not a particularly accurate result, and is only included here for reasons of completeness.

5. Conclusions

Electron beam irradiation of representative food samples on the Harwell electron linear accelerator HELIOS have been described. Dosimetry was established using an array of Faraday cups and Harwell Gammachrome YR perspex dosimeters.

In general, measurements of the radioactivity induced by the 20 MeV irradiations have been shown to agree well with the results of calculations. This agreement demonstrates that values of elemental concentrations in foods from standard tabulations, or from reasonable extrapolations of such tabulations, can be used for assessing the extent of any radioactivity induced by electron beam irradiation, although it is clear that in any future, similar experiments comprehensive chemical analysis of food samples should be carried out before irradiation. Useful limits have been placed on the induction of radioactivity by capture of neutrons produced in the samples themselves. The 20 MeV irradiations have also been used to simulate a gross malfunction of an electron beam irradiation plant, and the results show that for the maximum permitted dose of 10 kGy the specific activity after one day is only ~ 0.01 Bq g^{-1}.

The 10 MeV irradiations have been shown to have been effective in reducing the microbiological burden of the food samples by factors consistent with those found in previous measurements. As expected, no measurable radioactivity induced at 10 MeV was detected.

Acknowledgement—This work was carried out as part of the Corporate Research programme of AEA Technology.

*Randalls Road, Leatherhead, Surrey.

References

Allen P. D. and Chaudhri M. A. (1988) Photoneutron production in tissue during high energy bremsstrahlung radiotherapy. *Phys. Med. Biol.* **33,** 1017.

Anderson H. L. (Ed.) (1989) Physics Vade Mecum 2nd edn, Amer. Inst. Phys.

Barber W. C. (1958) Electrodisintegration of ^{9}Be and ^{12}C. *Phys. Rev.* **111,** 1642.

Becker R. L. (1983) *Absence of Induced Radioactivity in Irradiated Food, Recent Advances in Food Irradiation* (eds Elias P. S. and Cohen A. J.), p. 285. Elsevier Biomedical.

Berman B. L. (1974) Atlas of Photoneutron Cross-Sections obtained with Monoenergetic Photons, Lawrence Livermore Laboratory report UCRL 75694.

Bülow B. and Forkman B. (1974) Handbook on Nuclear Activation Cross-Sections, Tech. Rep. Ser. 156, IAEA.

Emmett M. B. (1975) The MORSE Monte Carlo radiation transport code system, Oak Ridge National Laboratory Report ORNL-4972.

Findlay D. J. S. (1989). Analytic representation of bremsstrahlung spectra from thick radiators as a function of photon energy and angle. *Nucl. Instr. Meth.* **A276,** 598.

Findlay D. J. S., Hawkes N. P. and Sené M. R. (1986) Photofission of ^{232}Th near threshold. *Nucl. Phys.* **A458,** 217.

Fleming R. F. (1985) An assessment of the induced radioactivity in foods using isotopic gamma-ray sources. *Trans. Am. Nucl. Soc.* **49,** 13.

Forkman B. and Petersson R. (1987) *Handbook on Nuclear Activation Data.* Tech. Rep. Ser. 273. IAEA.

von Janetschke P., Voigtländer K.-H. and Streuer H.-J. (1985), Lebensmittelhygienische Aspekte und Forderung im Zusammenhang mit der Lebensmittelbestrahlung. *Monatsch Veterinärmedizin* **40,** 600.

King J. D., Haslam N. H. and Parsons R. W. (1960) The gamma-neutron cross-section for ^{14}N. *Can. J. Phys.* **38,** 231.

Leboutet H. and Aucouturier J. (1985) Theoretical evaluation of induced radioactivity in food products by electron or X-ray beam sterilisation. *Radiat. Phys. Chem.* **25,** 233.

Lide R. R. (Ed.) (1990) *Handbook of Chemistry and Physics,* 71st edn. CRC Press, Boca Raton, FL.

Lynn J. E. (1980) HELIOS—The new Harwell electron linear accelerator and its scientific programme. *Contemp. Phys.* **21,** 483.

Matthews J. L. and Owens R. O. (1971) A single-difference bremsstrahlung unfolding technique for the measurement of high energy photonuclear cross-sections. *Nucl. Instr. Meth.* **91,** 37.

Miller A. and Jensen P. H. (1987) Measurements of induced radioactivity in electron- and photon-irradiated beef. *Appl. Radiat. Isot.* **38,** 507.

Mossel D. A. A. (1985) Irradiation—An effective mode of processing food for safety. *Proc. Int. Symp. Food Irradiation Processing,* Washington, D.C., 1985, p. 251. IAEA, Vienna.

Nascimento I. C. and Wolynec E. (1975) Nuclear excitation by electrons and positrons. *Nucl. Phys.* **A246,** 210.

Oikawa S. and Shoda K. (1977) Photoprotons from ^{44}Ca, ^{45}Sc and ^{46}Ti. *Nucl. Phys.* **A277,** 301.

Rogers F. and Whittaker B. (1970) The wholesomeness of irradiated food—the equation of induced activity, Harwell Report AERE R 6377.

Sharpe P. H. G. (1990) Radiation processing—The first thirty years, NPL News (National Physical Laboratory), No. 370, 25.

Whitaker B. (1990) A new PMMA dosimeter for low doses and low temperatures. *Radiat. Phys. Chem.* **35,** 699.

APPENDIX

Calculation of Induced Radioactivities

An expression for the bremsstrahlung flux produced by the aluminium scatter plate at the position of the food samples is

$$\frac{\mathrm{d}^2N}{\mathrm{d}k\,\mathrm{d}S}(E_0,k,r)=\frac{\mathrm{d}N}{\mathrm{d}k}(E_0,k)\times\sum_i\frac{a_i(E_0,k)}{\pi[b_i(E_0,k)\theta_b^2z_{rt}^2+\bar{\theta}_s^2(z_{rt}+z_{sr})^2+r_b^2+\bar{\theta}_d^2(z_{rt}+z_{sr})^2]}\times\exp-\frac{r^2}{[b_i(E_0,k)\theta_b^2z_{rt}^2+\bar{\theta}_s^2(z_{rt}+z_{sr})^2+r_b^2+\bar{\theta}_d^2(z_{rt}+z_{sr})^2]}$$

where $\mathrm{d}^2N/\mathrm{d}k\,\mathrm{d}S$ is the number of photons per MeV per cm^2 of energy k per incident electron at a distance r from the axis of the incident electron beam of energy E_0, a_i and b_i are the parameters of a sum-of-gaussians expansion of the angular distribution of bremsstrahlung from the scatter plate, θ_b is the characteristic bremsstrahlung angle $\theta_b=m_ec^2/(m_ec^2+E_0)$ where m_e is the electron mass, θ_s^2 is the mean square multiple scattering angle for the 0.1 mm electron beam window upstream of the scatter plate, r_b and θ_d^2 are respectively the $1/e$-radius and the mean square angular divergence of the electron beam before it hits the electron beam vacuum window, and z_{rt} and z_{sr} are the distances between the scatter plate and the samples and the scatter plate and the preceding electron beam vacuum window respectively. On the basis of this expression the yield y_j^γ of nuclide j in the sample is

$$y_j^\gamma=\frac{f_j\rho}{A_j}N_\mathrm{A}\int_{k_{\mathrm{th}j}}^{E_0}\frac{\mathrm{d}^2N}{\mathrm{d}k\,\mathrm{d}S}(E,k,\bar{r})\sigma_j(k)\,\mathrm{d}k$$

where y_j^γ is the number of atoms of nuclide j per cm^3 per electron, f_j is the fraction by weight of the parent of nuclide j with atomic weight A_j and cross-section σ_j in the sample contributing to the bremsstrahlung-induced activity, ρ is the sample density, N_A is Avogadro's number, and $\bar{r}$ is a suitably averaged radial coordinate. A convenient analytic expression for the bremsstrahlung spectrum per incident electron from the scatter plate $\mathrm{d}N/\mathrm{d}k(E_0,k)$ must be taken from Findlay (1989) in which the integrated-over-angle intrinsic bremsstrahlung intensity cross-section is taken as a linear function of k

$$\frac{k}{Z^2}\frac{\mathrm{d}\sigma}{\mathrm{d}k}=a\left(1-b\frac{k}{E}\right).$$

For aluminium, $a\simeq 13$ mb and $b\simeq 0.93$.

By assuming the same linear representation of the integrated-over angle intrinsic bremsstrahlung intensity cross-section, an expression for the yield induced by bremsstrahlung produced in the samples themselves is

$$y_j^\mathrm{s}=N_\mathrm{A}^2\int_0^{\min(\rho X,(E_0-k_{\mathrm{th}j})/[\mathrm{d}E/\mathrm{d}(\rho x)])}\frac{1-\exp[-(\mu/\rho)(\rho X-\rho x)]}{(\mu/\rho)}\times\left\{\int_{k_{\mathrm{th}j}}^{E_0-x\,\mathrm{d}E/\mathrm{d}x}\sum_i f_i\frac{Z_i^2}{A_i}\frac{a_i}{k}\left(1-b_i\frac{k}{E_0-\rho x\,\mathrm{d}E/\mathrm{d}(\rho x)}\right)\times f_j\frac{1}{A_j}\sigma_j(k)\,\mathrm{d}k\right\}\mathrm{d}(\rho x)$$

where y_j^s is the number of atoms of nuclide j per electron, N_A is Avogadro's number, E_0 is the incident electron energy, μ/ρ is the photon mass attenuation coefficient, ρX is the sample thickness (g cm^{-2}), $\mathrm{d}E/\mathrm{d}(\rho x)$ is the electron stopping power, f_i' is the fraction by weight of element Z_i, A_i contributing to bremsstrahlung in the sample, a_i and b_i are parameters of the straight-line representation of the corresponding integrated-over-angle intrinsic bremsstrahlung intensity cross-section, f_j is the fraction by weight of the parent of nuclide j with atomic weight A_j and cross-section σ_j contributing to the photonuclear yield. The sample thickness was assumed to be 2.5 cm.

The yield due to electronuclear reactions can be calculated using the concept of virtual photons to relate the electron-induced reaction cross-section to the corresponding photon-induced cross-section (Barber, 1958). An expression for the electron-induced yield in the sample is then

$$y_j^\mathrm{y}=\frac{f_j}{A_j}N_A\int_0^{\min(\rho X,(E_0-k_{\mathrm{th}})/[\mathrm{d}E/\mathrm{d}(\rho x)])}\int_{k_{\mathrm{th}}}^{E_0-\rho x\,\mathrm{d}E/\mathrm{d}(\rho x)}\frac{\mathrm{d}N}{\mathrm{d}k}[Z,E_0-\rho x\,\mathrm{d}E/\mathrm{d}(\rho x),k]\sigma_j(k)\,\mathrm{d}k\;\mathrm{d}(\rho x)$$

where y_j^y is the number of atoms of element j per electron f_j, ρ, A_j, N_A, ρX, $\mathrm{d}E/\mathrm{d}(\rho x)$ and E_0 are as above, $\mathrm{d}N/\mathrm{d}k(Z,E,k)$ is the virtual photon spectrum as a function of virtual photon energy k for electron energy E and atomic number Z as given, for example, by Nascimento and Wolynec (1975). A convenient expression (Matthews and Owens, 1971) for the virtual photon spectrum $\mathrm{d}N/\mathrm{d}k(Z,E,k)$ is

$$\frac{\mathrm{d}N}{\mathrm{d}k}(E,k)=\frac{1}{137\pi}\frac{1}{k}\times\left\{\frac{E^2+E'^2}{p^2c^2}\ln\left(\frac{EE'+pp'c^2+m_e^2c^4}{m_ec^2(E-E')}\right)-2\frac{p'}{p}\right\}$$

where $p^2c^2=E^2-m_e^2c^4$, $p'c=pc-k$ and $E'^2=p'^2c^2+m_e^2c^4$. This assumes electric dipole transitions only and neglects Coloumb distortion, but the Coulomb distortion only amounts to, at most an $\sim 10\%$ effect for materials up to $Z\simeq 20$ at the present energies.

Memoranda/Mémorandums

Food safety aspects relating to the application of X-ray surveillance equipment: Memorandum from a WHO meeting*

Inspection of food-containing cargoes using X-rays is safe since no detectable radioactivity will be induced in the foodstuffs provided that an energy level of 10 MeV and a dose of 0.5 Gy are not exceeded.

Introduction

Many countries have regulations permitting the irradiation of foodstuffs. In most cases, these regulations conform to the Codex General Standard for Irradiated Foods (*1*), and in particular specify that X-rays used for this purpose should be generated from machine sources operating at or below an energy level of 5 mega-electron-volts (MeV). This limit has been chosen in order to stay well below the energy level where significant induction of radioactivity in the irradiated food may be expected.

Ionizing radiation is used not only to accomplish an effect on food, but also in connection with process and quality control (e.g., detection of the level of filling in cans and of foreign-bodies in containers) and in connection with the use of X-ray surveillance equipment. WHO has recently been informed of new technological developments that have made it possible to use higher energy X-ray systems for the examination of large cargo containers and cargo vehicles to detect the presence of contraband such as illegal drugs, explosives and guns. Some Member States of WHO have already expressed interest in the use of such surveillance equipment. However, for penetrating large cargo containers, these systems operate with X-ray energies of over 5 MeV.

Although there may be considerable advantages in using this new technology in combating terrorism, etc., countries may be hesitant in allowing the use of such equipment on cargoes containing food because the energy level is in excess of that specified for food irradiation by the Codex Alimentarius Commission.

It was for this reason that WHO, in cooperation with IAEA, convened a meeting to seek international consensus on the food safety aspects arising from the use of high-energy X-ray surveillance systems. All companies known to WHO as developers or manufacturers of X-ray surveillance equipment were invited to participate. Their representatives presented technical information on such equipment and surveillance systems at the meeting.

The objectives of the meeting were:

— to investigate the usefulness of inspecting food-containing cargoes with the help of ionizing radiation;
— in the event of an affirmative answer, to define the parameters (energy-level and dose) necessary for large cargo surveillance with X-rays; and
— to consider possible health consequences from exposing food to X-rays with energies greater than 5 MeV and an absorbed dose in the range of 0.5 gray (Gy), in relation to induction of radioactivity; toxicological, nutritional and sensory considerations; and microbiological considerations.

* This Memorandum is based on the report of a WHO Consultation, convened in cooperation with the International Atomic Energy Agency (IAEA), which met in Neuherberg/Munich, Federal Republic of Germany on 13–17 November 1989. The participants were A.M.I. Alsayyed, Doha, Qatar; K.J. Dale, London, England; J.F. Diehl, Karlsruhe, Federal Republic of Germany; J. Farkas, Budapest, Hungary (*Rapporteur*); M. Frissel, Bilthoven, Netherlands; H. Fröhlich, Frankfurt, Federal Republic of Germany; J.H. Hubbel, Gaithersburg, MD, USA (*Chairman*); J.R. Lujan, Mexico DF, Mexico; and G. Pauli, Washington, DC, USA. *Secretariat*: K.W. Bögl, Berlin (West); A. Brynjolfsson, Wageningen, Netherlands; F.K. Käferstein, WHO (*Secretary*); A.-M. Schmitt-Hannig, IAEA; R.B. Singh, London, England; and H. Stiff, WHO. *Joint FAO/WHO Food Standards Programme*: E. Casadei, FAO, Rome, Italy. In addition, companies interested in X-ray surveillance equipment were represented by G. Geus and C. Koch, Wiesbaden, Federal Republic of Germany; C.T. Blunden and G. Bennet, Bristol, England; and C.S. Nunan, Palo Alto, CA, USA. Requests for reprints should be sent to Dr F.K. Käferstein, Food Safety Unit, Division of Environmental Health, World Health Organization, 1211 Geneva 27, Switzerland. A French translation of this Memorandum will appear in a later issue of the *Bulletin*.

Reprint No. **5075**

Memorandum

Cargo inspection

Is inspection with X-ray surveillance equipment useful?

A major commitment of the customs authorities the world over is the fight against illegal trafficking in contraband such as drugs and arms. There are at least two factors which have an important bearing on the efficiency with which this task is performed: (1) the need to unpack and repack cargo items; and (2) the huge volume of cargoes at the present time and the increases anticipated in the future.

At Hamburg port, for instance, container traffic increased by 11.7% in 1988 to a total of 1.6 million containers. Dover and Southampton ports together handle approximately 20 tonnes of food per minute every day, which amounts to some 9.5 million tonnes/year. Throughput in the State of Qatar is some 20–40 trailers of foods each day. Similar considerations apply to air cargo. At Frankfurt International Airport, for example, 2.2 million individual consignments are handled annually by the customs authorities; an expansion by about 33% is expected by the year 2000. All the above figures are likely to increase with the anticipated rise in world food trade.

Control procedures for detecting and preventing contraband fall into a number of categories, such as the use of (1) conventional manual control; (2) dogs for detecting drugs and explosives; (3) chromatographic, spectroscopic and related methods; and (4) X-ray surveillance.

The advantage of the first three of these methods is the immediate provision of incriminating evidence, thus permitting direct assessments to be made. A disadvantage of the second and third methods is that these are highly specialized techniques and therefore of limited general applicability. Also, for biological reasons, dogs cannot repeatedly provide satisfactory results over extended periods. The most important drawback of all three methods is that they are time-consuming and labour intensive and, consequently, do not permit a high throughput of goods generally and large cargo containers in particular.

The fourth method, X-ray surveillance, is a rapid and efficient tool for the systematic and serial inspection of cargoes. However, X-ray surveillance systems currently in use operate at 140 kilovolts (i.e., energy levels[a] up to 0.14 MeV); because of this technical limitation, present systems allow for the inspection of small cargoes only. It is understood that recent developments, using surveillance equipment with X-ray energy levels of up to 10 MeV, will enable large cargo containers to be screened without the need for opening the container and unpacking the goods.

This new technique will therefore facilitate the checking of large volumes of bulk consignments such as perishable goods (e.g., fresh food, flowers, etc.), textiles and leather goods without the need for unpacking. This is a particularly important consideration in view of the extraordinary inventiveness of smugglers in thinking up places and means of concealment. Perishable goods are an example in point; because of the known difficulties in handling such cargoes (time constraints, financial penalties), these goods are being used, increasingly, to conceal contraband, mostly drugs. It should be noted that the use of high-energy X-ray equipment requires experienced personnel trained in image interpretation and in its safe operation (for details, see Annex page 301).

Any development which facilitates rapid screening of large cargo containers will be advantageous to the customs and other control authorities. However, the technical feasibility and health consequences of such high-energy surveillance systems are issues that are discussed below.

Parameters necessary for X-ray surveillance of large cargo containers

Energy levels. X-ray surveillance of large cargo items with thicknesses of the order of 2.5 m of water equivalent, or 30 cm of steel, is not possible without increasing the penetrating power of the X-ray beam. The penetrating power can be increased only by increasing the energy levels from those at present used for luggage inspection, typically up to 0.14 MeV, to energy levels of the order of 5 to 10 MeV.[b]

For successful imaging, including use of various kinds of image enhancement techniques, the maximum tolerable attenuation of the primary X-ray beam in traversing the cargo unit appears to be between 10^{-4} and 10^{-5}. The 10^{-4} figure comes from presentations at this Consultation by representatives of companies producing fan-beam, moving-cargo high-energy X-ray surveillance equipment. The 10^{-5} figure was inferred from published information on rocket-motor flaw detection in 50 cm of steel using 16 MeV X-rays (*2*).

[a] For the purpose of this report, the term "energy level" is defined as the maximum photon energy producible by the X-ray source.

[b] One company described X-ray surveillance equipment using maximum energy levels of 8 MeV; another company demonstrated images derived from equipment operating at energy levels between 6 MeV and 10 MeV; and a third company suggested the possibility of using energy levels greater than 10 MeV.

The penetrating power of the X-ray beam does not increase indefinitely with increasing photon energy. There is a minimum in the attenuation cross-section vs. photon energy, above which the X-ray beam becomes less penetrating (*3*). For carbon, this minimal attenuation energy is 55 MeV, but drops to 8 MeV for copper and to 3.5 MeV for lead.

Another factor to be considered is the contribution to the attenuation from photonuclear interactions (*4*) in the 6–30 MeV region which accounts for only 2–6% of the total attenuation, but which can be a major mechanism for inducing radioactivity in the cargo material. This consideration is discussed in more detail below.

Dose levels. Information on the dose requirements for imaging with multi-MeV photons appears to be currently available only from commercial developers of such equipment.

For imaging with a cone-beam and a two-dimensional imaging screen (stationary cargo), the presentation by a representative of a developer of this type of equipment highlighted the need for a dose of 0.05 Gy at the surface of the cargo nearer to the X-ray source. This would imply a dose at the detector side of the cargo of 0.05×10^{-4} Gy (i.e., 5×10^{-6} Gy) required by the detector system to produce an acceptable image.

For imaging with a fan-beam (moving cargo) facility using two beams at right angles, a much lower dose may be possible (in this context, a dose as low as 0.00025 Gy at the source side was quoted by one producer).

To allow for flexibility, for overlap of the exposures in some systems, for sufficient resolution, and for the need to re-examine cargoes in some instances, the Consultation considered a maximum dose of 0.5 Gy absorbed by the food.

Possible health consequences

Exposure of food to X-rays with energy levels >5 MeV and a maximum dose of 0.5 Gy

Induction of radioactivity. Several possibilities exist to induce radioactivity in food. The induction depends on an interaction between X-ray photons or neutrons with atoms in the food. Most interactions of this kind do not lead to the induction of radioactivity.

One type of interaction produces radioactive isomers. Energy from a photon is absorbed by an atom and afterwards emitted as radiation. Neutrons may be emitted following interactions of photons with atoms in the food (e.g., deuterium), or from outside sources (for example, as used in a thermal neutron detection scanning device). The absorption of a neutron by an atom may also induce activity. Electrons induce radioactivity primarily by indirect means; photons are created (*Bremsstrahlung*) as the electrons strike the target material. These photons, in turn, interact with the nucleus of the atom in photonuclear reactions. Many of these reactions have threshold energies below which reactions do not occur. Thresholds are always dependent on the isotope and the type of reaction. All these physical processes are well known and documented and amenable to calculation. Results of such calculations are reported by Becker (*5,6*) and by Leboutet & Aucoutuviev (*7*).

Based on such calculations, the Consultation recognized that high-energy radiation can induce radioactivity in any absorbing medium, such as food. For example, one can calculate that even natural background radiations (e.g., cosmic rays) induce radioactivity in food. The factors affecting the radioactivity include the type of radiation (electromagnetic, probability of induced electron or neutron), the energy of the radiation, and the particular elements found in the food. These factors can also interact; for example, high-energy X-rays can induce reactions that produce neutrons, leading to further reactions caused by the neutrons.

Experimental studies that are relevant to determine the effects of low-dose/high-energy X-rays on food are usually not designed to determine induced radioactivity at the combinations of energy level, dose, and time after exposure that would be used in surveillance systems. However relevant experimental data are available from studies designed to evaluate the use of activation analysis and the application of X-rays and electrons in food irradiation and medical uses at energy levels up to 24 MeV and at doses up to 50 kGy. Such studies, both theoretical and experimental, can be used to extrapolate downwards to a lower dose such as that of 0.5 Gy considered by the Consultation for surveillance systems. These studies show no evidence that detectable levels[c] of radioactivity would be induced at these lower doses.

In light of the large variations of background radioactivity in food that are of no concern, the Consultation concluded that radioactivity below the detection limit is also of no concern. A criterion of no detectable, induced radioactivity may be more strict

[c] All foods contain radioactivity, usually at levels in the range of 30–300 becquerel/kg. The amount of radioactivity in any specific food varies, depending on its elemental composition. The amount of increased radioactivity that can be measured is typically about 1% of the natural background in the food. For the purpose of this report, the Consultation considered this level to be the detection limit.

than necessary. However, present-day technology is capable of producing X-ray surveillance equipment which does not induce detectable amounts of radioactivity. Therefore, such a criterion provides a sufficient margin of safety to eliminate the need for considering cumulative effects of repeated X-ray surveillance inspections or occasional deviation from intended conditions of use due to human error.

Toxicological, nutritional and sensory considerations. The Consultation considered the question of whether high-energy X-ray surveillance of food-containing cargo might cause chemical changes of toxicological or nutritional concern, or changes in the sensory quality of food. The conclusion was that, at the considered radiation dose of 0.5 Gy for X-ray cargo inspection, radiation-induced chemical changes in foods are so minute that no toxicological risks, losses of nutrients or changes in sensory quality can be foreseen. The dose level that might require consideration of such risks or changes is considerably greater than that needed for surveillance; therefore, even repeated inspections of the same cargo would not be of concern.

Microbiological considerations. The microbiological safety of irradiated foods has been investigated in many laboratories in relation to food preservation by ionizing radiation, and was a subject of discussion at several international meetings of experts, including the Joint FAO/IAEA/WHO Expert Committee on Wholesomeness of Irradiated Food (*8*). The conclusion of these reviews was that the microbiological safety of irradiated food is fully comparable with that of foods preserved by other acceptable preservation methods.

Regarding the energy levels of X-ray surveillance equipment which are higher than those at present permitted for food preservation, the Consultation concluded that the events following the primary interactions, including chemical and radiobiological effects, are the same and are independent of the different proportion of various primary energy absorption processes during interaction of X-rays with matter as a function of increasing photon energies. Thus, in principle, the same main questions which have been scrutinized in the past in relation to microbiological safety of radiation-preserved food may be considered also for high-energy X-ray surveillance of food. However, the much lower dose requirement of the latter technique should be taken into consideration. Regarding dose requirement for selective changes in the composition of the microflora and for changes in the diagnostic characteristics of microorganisms, and considering the fact that nothing of significance has been found regarding radiation-induced mutants even at the dose levels of food preservation by irradiation, the Consultation concluded that no microbiological hazard will arise from the use of the proposed X-ray surveillance systems.

Conclusions

The Consultation concluded that of all the issues discussed, only the induction of radioactivity may be of concern regarding the potential effects of health. Evaluation of the likelihood of inducing radioactivity in food has mostly been based on theoretical calculations because the X-ray surveillance systems currently under consideration are not capable of producing detectable levels of activity.

Calculations applied to the different possibilities can be quite complex. It is not essential to make precise calculations, however, if a sufficient safety margin is built in to the deliberation. This condition is met when no detectable radioactivity is induced in foodstuffs.

The Consultation concluded, on the basis of available evidence, that no detectable radioactivity will be induced in foodstuffs when an energy level of 10 MeV and a dose of 0.5 Gy are not exceeded. The safety of the food will not be affected as a consequence of such exposure.

However, this conclusion is not intended to preclude other safe surveillance systems designed to operate at a higher energy level or dose. In such cases, assurance should be provided that, at the point of consumption, food would not contain a measurably detectable amount of induced radioactivity.

Acknowledgements

The Consultation was supported by a grant from the United Kingdom government.

References

1. **Joint FAO/WHO Food Standards Programme.** Codex general standard for irradiated foods and recommended international code of practice for the operation of radiation facilities used for the treatment of foods. *Codex alimentarius*, Vol. XV, 1st edition. Rome, Codex Alimentarius Commission, 1984.

2. **Bakke, T.O.** (quoting T.E. Kirchner, American Science and Engineering Inc., Cambridge, MA), Giant scanner inspects rocket motors. *Popular science*, **224**: 95 (1984).
3. **Hubbell, J.H. et al.** Pair, triplet, and total atomic cross-sections (and mass attenuation coefficients) for 1 MeV–100 GeV photons in elements Z=1 to 100, *J. phys. chem. ref. data*, **9**: 1023–1147 (1980).
4. **Fuller, E.G. & Hayward, E.,** ed. Photonuclear reactions Stroudsburg, PA, Dowden, Hutchinson & Ross, 1976.
5. **Becker, R.L.** Radioactivity induced in foods by 10 MeV electron irradiation. Natick, MA, US Army Natick Research and Development Command, 1977.
6. **Becker, R.L.** A determination of the radioactivity induced in foods as a result of irradiation by electrons of energy between 10 and 16 MeV. Natick, MA, US Army Natick Research and Development Command, 1979.
7. **Leboutet, H. & Aucouturier, J.** Theoretical evaluation of induced radioactivity in food products by electron and X-ray beam sterilization. *Radiat. phys. chem.*, **25**: 233 (1987).
8. WHO Technical Report Series No. 659, 1981 (*Wholesomeness of irradiated food*: report of a Joint FAO/IAEA/WHO Expert Committee).

Annex

Operational radiological safety aspects

Electron linear accelerators are being used throughout the world in increasing numbers in a variety of important applications. Foremost among these is their role in the treatment of cancer with both photon and electron radiations in the energy range 4–40 MeV. To a greater extent linear accelerators are replacing Co^{60} sources and betatrons in medical applications. Commercial uses include non-destructive testing by radiography, food preservation, product sterilization and radiation processing of materials such as plastics and adhesives. Scientific applications include investigations in radiation biology, radiation chemistry, nuclear and elementary-particle physics and radiation research.

Guidelines and standards on the radiological safety aspects of the operation of such accelerators have been developed on a national[a,b] and international[c] basis.

In view of the rapidly growing number of cargo container shipments throughout the world, a new field of application for linear accelerators with photon energies of about 10 MeV has been established for X-ray surveillance of large containers.

In principle, the same registration, licensing and inspection procedures established by the appropriate regulatory authority apply as for all linear accelerators operating in the same energy range. In countries where a proper radiation protection infrastructure is not available, the Consultation suggests that the manufacturer should notify the IAEA. However, the responsibility for protection of personnel, facilities, the public and the environment from all types of hazards related to linac (linear accelerator) operations must rest with the management of the organization using these systems. Under its direction, a safety unit should be established and a safety programme appropriate to the special needs of the application should be developed and implemented.

A radiation safety programme should be developed in coordination with the facility's overall safety programme, and in compliance with national, regional and local requirements. Recommendations of international organizations such as IAEA, the International Commission on Radiological Protection (ICRP), the International Commission on Radiation Units and Measurements (ICRU), the International Electrotechnical Commission (IEC) and the Commission of the European Communities, as well as national commissions, should be considered in the development of this programme.

[a] **United States Atomic Energy Commission.** *Safety guidelines for high-energy accelerator facilities.* Washington DC, National Accelerator Committee, USAEC Division of Operational Safety, 1967 (see the latest version).

[b] **United States Atomic Energy Research and Development Administration.** *Operational safety standards.* Washington DC, AECM Section 0550, USERDA (periodically revised).

[c] IAEA Technical Report Series No. 188 (*Radiological safety aspects of the operation of electron linear accelerators*). Vienna, International Atomic Energy Agency, 1979.

FOOD AND AGRICULTURE ORGANIZATION
OF THE UNITED NATIONS

INTERNATIONAL
ATOMIC ENERGY AGENCY

WORLD HEALTH
ORGANIZATION

INTERNATIONAL CONSULTATIVE GROUP ON FOOD IRRADIATION
(ICGFI)

SECRETARIAT: JOINT FAO/IAEA DIVISION OF NUCLEAR TECHNIQUES IN FOOD AND AGRICULTURE, INTERNATIONAL ATOMIC ENERGY AGENCY, WAGRAMERSTRASSE 5, P.O.BOX 100, A-1400 VIENNA, AUSTRIA
TELEPHONE: + 43 1 2600 21638, FACSIMILE: + 43 1 26007, E-Mail: Official.Mail@iaea.org INTERNET: http://www.iaea.org/icgfi/

THE DEVELOPMENT OF X-RAY MACHINES FOR FOOD IRRADIATION

(PROCEEDINGS OF A CONSULTANTS' MEETING)

Vienna, Austria
16-18 October 1995

ICGFI is an International group of experts designated by Governments to evaluate and advise on global activities of food irradiation.

tantalum and gold) can produce photo-neutrons if the electron energy is raised to 10 MeV. However, limiting the energy to 7.5 MeV would prevent production of photo-neutrons in gold converters and limit the photo-neutrons produced in tungsten and tantalum converters to an insignificant number.

The Consultation also concluded that the radiation safety requirements for machines operating at 7.5 MeV would not be different from those imposed upon machines operating at lower energies. It was also concluded that existing dosimetry methods for X-ray processing would be appropriate for machines operating at 7.5 MeV.

Efficiency

If the commercial application of radiation processing using X-rays is to be a success then the technology must be at least as efficient in utilizing energy, and so achieving throughput of product, as existing methods (primarily radionuclide facilities). The various factors which affect energy efficiency were considered (photon utilization, conversion efficiency, self-absorption) and it was concluded that an overall efficiency of approximately 8% could be achieved using 7.5 MeV compared to 4% which can be achieved using X-rays generated from machines with a maximum energy of 5 MeV. Thus, the efficiency achievable at 7.5 MeV is comparable to that achieved in radionuclide facilities.

Economics

An economic model was used to investigate how various parameters (the dose required, the beam power and the energy generated by the machine) affected the cost of the process. The use of 7.5 MeV was found to be more cost effective than using X-rays generated from machines with a maximum energy of 5 MeV. For example, at a dose of 2.5 kGy, using a beam power of 100 kW, treatment with a 5 MeV machine would cost US$ 52.5 per tonne of material, compared to US$ 35 per tonne using 7.5 MeV.

CONCLUSIONS

It was concluded that X-ray machines for food irradiation with energy up to 7.5 MeV can be used without any concern about induced radioactivity but would be a satisfactory, efficient and cost effective addition to other radiation sources available for food processing.

relatively short time, but also to provide the food industry with different options of irradiation facilities. An example of such a situation is the treatment of Chilean grapes to satisfy quarantine regulations for importation in the U.S.A. Over 100,000 tonnes of grapes have to be treated within the space of a few months. Table 1 lists food items which could be advantageously treated by X-rays. Thus, efficient, economical, and high capacity X-ray machines could make an important contribution to food irradiation under specific circumstances and would reduce the burden on other types of radiation sources.

Table 1. Food and agricultural commodities which may be more efficiently treated by X-rays.

1. Seasonal Fruits to Overcome Quarantine Barriers
   Grapes, mangoes, papaya, carambola, lychees, rambutan, etc.

2. Prepackaged, Fresh and Frozen Food of Animal Origin
   Poultry, meat, seafood, processed food products (especially for bulky products for in-line irradiation facilities).

3. High-Value Stored Food Products
   Dried fish, dried meat, dried fruits and tree nuts, cocoa beans, etc. (especially those which have high volume and are seasonal in production)

4. Cut-Flowers
   Various types of prepackaged cut-flowers/foliages to overcome quarantine barriers

It is anticipated that there will be a significant demand for large quantities and a variety of radiation sources for food processing in the near future for the following major reasons:

(i) Reduction of Foodborne Diseases. The increasing awareness of the risks from foodborne diseases and the demand for microbiologically safe food by the consumer will lead to a wider use of irradiation as a cold pasteurization process of foods, especially those of animal origin.

(ii) Replacement of Fumigation of Food. The global phasing out of methyl bromide (the most widely used fumigant to control insect infestation of food and agricultural commodities) under the Montreal Protocol[1] by the year 2000 will have an important impact on trade in food and agricultural commodities which have to be treated to overcome insect problems. Irradiation is likely to replace the use of methyl bromide for a wide variety and large quantities of fresh and dried fruits and tree nuts, especially to overcome quarantine barriers.

[1] An international treaty for the regulation of ozone depleting substances worldwide and under the auspices of the United Nations Environmental Programme.

2.1.1. What is significant induced radioactivity?

The most sensitive analytical measurements can detect radioactivity at a level of 1% of the natural radioactivity in food. This limit, together with an additional safety factor of 10, allows induced activity to be defined as significant if it is more than 1/1000 of the natural background in food. As shown above, there can be considerable variation in the natural radioactivity in food, but assuming an average value of 200 Bq/kg, induced radioactivity would therefore not be significant below 0.2 Bq/kg.

2.2. RADIOACTIVITY INDUCED BY ELECTRONS, GAMMA-RAYS, AND X-RAYS

Presently, the Codex Alimentarius Commission permits that food be exposed to gamma-rays from ^{60}Co and ^{137}Cs, to fast electrons less than 10 MeV, and to X-rays less than 5 MeV. The maximum average dose is 10 kGy. These radiation sources do not induce measurable radioactivity in the food, and theoretical calculations show that any induced activity is actually many orders of magnitude less than the limits defined above. The theoretical calculations show that there are three main pathways for induction of radioactivity in food: (i) isomeric activation; (ii) photo-nuclear activation; and (iii) neutron activation.

The analysis further shows that in the case of irradiation by 10 MeV electrons and by 5 MeV X-rays, the neutron activation, although insignificant, is larger than the activation produced by the other two major pathways, and that the neutron activity produced by 5 MeV X-rays is in the order of 60 times greater than that produced by 10 MeV electrons. Therefore, neutron activation is discussed in more detail below.

2.2.1. Neutron Activation

The threshold energy for the gamma-neutron, (γ, n), reaction is well above the 10 MeV energy limit for all the major isotopes in food. Thus, the threshold in the major isotopes of carbon, oxygen and nitrogen are 18.72 MeV (^{12}C), 15.67 MeV (^{16}O) and 10.55 MeV (^{14}N) respectively. However, a few isotopes have low photo-neutron thresholds; namely 2.225, 4.85 and 4.15 MeV in deuterium (hydrogen-2 (^{2}H)), ^{13}C and ^{17}O isotopes respectively. The concentration of these isotopes is low and the isotopes ^{1}H, ^{12}C and ^{16}O, which are produced when the neutron is ejected, are stable. Many of the trace elements and contaminants in food also have thresholds slightly below 10 MeV. The cross-sections (that is, the probabilities for the processes) are small and for most isotopes (except deuterium) increases in this range approximate to the third power of the excess energy of the electrons above the (γ, n) threshold (that is, $\sim (E - E_{th})^3$).

The neutrons emitted in these processes usually have initially an energy of a few MeV, but they will gradually be slowed down by collisions with the atoms of food and 'thermalized'. Some of the neutrons will escape the food and be absorbed in the conveyor and walls of the irradiation chamber, but some will be absorbed in the food. In a model food of average composition, about 89.4% of neutrons absorbed in the food will be absorbed in hydrogen to reform the stable isotope deuterium, about 8.5% will be absorbed in the ^{14}N to form the nearly stable and stable isotopes ^{14}C and ^{15}N respectively, about 1.1% will be absorbed in chlorine-35 (^{35}Cl) to form the nearly stable isotope ^{36}Cl and 3000 times less of the isotope sulphur-35 (^{35}S) with half-life of 86.7 days, about 0.54% will be absorbed in ^{39}K to form the nearly stable isotope ^{40}K, and about 0.17% will be absorbed in ^{12}C to form the stable isotope ^{13}C. The remaining will be

(ii) tantalum with thresholds at 6.6 MeV in ^{180}Ta (0.012%) and 7.6 MeV in ^{181}Ta (99.988%).

(iii) gold with a threshold at 8.1 MeV in ^{179}Au (100%).

Thus increasing the energy of X-rays above 7.5 MeV would result in increased cross-section (probability) of neutron production in the X-ray converter and, consequently, in possible induction of radioactivity in the irradiated food.

2.2.2. Control of energy

Direct current (DC) electron accelerators usually have a well defined maximum energy. Linear accelerators, when well tuned and properly operated, also have a rather well defined maximum energy. The width of the electron energy spectrum at half maximum may then be less than 0.25 MeV. However, if the accelerator is not tuned for optimally small energy distribution the width may be much greater. As in all measurements, the variations in parameters must be taken into account. The (γ, n) thresholds for copper-63 (^{63}Cu) is 9.91 MeV and ^{62}Cu has a half life of 9.8 minutes; the threshold for ^{65}Cu is 10.84 MeV and ^{64}Cu has a half life of 12.9 hours. These isotopes can be used to determine the 10 MeV electron energy. The positron radioactivity increases ~$(E - E_{th})^3$ for each isotope, and then can easily be used to define the energy within 0.1 MeV by measuring how the activity increases with energy. Coincidence measurements of the two 0.511 MeV quanta produced in the decay of the positrons can be used to reduce or eliminate the background. Such measurements can therefore detect minute radioactivity produced in copper wires exposed to the electron beam. Similarly, the energy and the design of the converter area in an X-ray facility can be controlled by having a competent laboratory measure the neutron induced radioactivity in a gold foil placed at the center of a 15 cm x 15 cm x 15 cm water phantom in the sample area. If calibrated foils indicate a fluence of more than 3.5×10^5 neutrons per kGy dose in the sample, the energy is too high.

2.3. CONCLUSIONS

The review of the literature and the analysis presented here indicate that radiation processing with X-rays up to 7.5 MeV can be used without any concern about induced radioactivity, provided special care is taken in the design of the X-ray converter so as to eliminate significant neutron production in the converter.

3. ENERGY CONVERSION EFFICIENCY

There are three main factors governing overall electron-power utilization efficiency: photon-power utilization, conversion efficiency in converter, and self-absorption correction.

(i) photon-power utilization: as in case of ^{60}Co-gamma rays, not all of the photons produced in the X-ray converter are absorbed in the products being irradiated. Electromagnetic energy is attenuated in matter following exponential laws. Consequently, the fact that some part of the impinging energy leaves the products cannot be avoided.

3.3. SELF-ABSORPTION

In a properly designed X-ray converter only 10 to 20% of the photons produced are absorbed in the convertor itself.

3.4. OVERALL ELECTRON-POWER UTILIZATION

The following (Table 2) combines the components discussed above.

Table 2. Factors contributing to energy conversion efficiency.

	5 MeV (%)	7.5 MeV (%)
Photon utilization	30	40
Conversion efficiency	14	22
Self-absorption correction	80	90
Overall	4	8

In conclusion, an overall energy conversion efficiency (ie electrons to photons absorbed) of 4% at 5 MeV is achieved; increasing this limit to 7.5 MeV can increase efficiency up to 8%. Taking into account the power in the electron beam of commercially available accelerators, and the conversion efficiencies to X-rays of 4 - 8%, it is concluded that X-ray radiation processing facilities of throughput capacities comparable to isotope facilities are available.

4. ECONOMICS OF HIGH ENERGY, HIGH CAPACITY X-RAY MACHINES

Modern accelerators are made to industrial standards and specifications and such machines at high power and high electron energy are operated on an industrial scale at many installations and the records for availability are very good. Trained and qualified personnel are needed to maintain such facilities; however, with the help of modern computer technology and by the design and engineering of the components for greater reliability such accelerators are now typically operated by a trained staff who do not need a professional background in accelerator, high-voltage and other techniques.

Significant progress has been made in the development of X-ray machines for radiation processing. Ten years ago high power machines with average power up to 150 kilowatt (kW) had a maximum energy of 4.5 MeV. More recently, this energy has been increased to 5 MeV by at least two manufacturers. New radiofrequency (RF) technologies with energies up to 10 MeV have been demonstrated at power levels up to 100 kW and are operating commercially at 50 kW. The life time cumulative availability of the latest of these machines matches the established reliability of DC machines. Plans are underway to extend these power levels to 200 kW at the 10 MeV level.

The opportunity to reevaluate the maximum energy of X-ray machines may be done with the knowledge that accelerators are available with electron energies above 5 MeV. It can be concluded that technologies are now a matter of choice between DC and RF machines and the choice will be driven by fundamental economic issues.

6. RADIATION SAFETY

6.1. SAFETY PHILOSOPHY

The radiation safety considerations of X-ray machines with energies between 5.0 and 7.5 MeV will not be appreciably different from those with energies less than 5.0 MeV.

6.2 SHIELDING

Increasing the energy above 5 MeV will require the radiation shield thickness (concrete) to be increased by a few centimeters. However, the procedures for checking the effectiveness of the shield are no different from those used with X-ray machines with energies less than 5 MeV.

6.3. INDUCED INACTIVITY

Increasing the energy to 7.5 MeV will not significantly increase the radioactivity in the food or the concrete shielding material, the structural materials of the machine or the conveyor system.

An X-ray converter made of gold will not produce any photo-neutrons, as the threshold for gold is 8.1 MeV. For tungsten with a threshold energy of 6.2 MeV and tantalum with a threshold energy of 6.6 MeV only a few photo-neutrons will be produced. Therefore, the neutron induced activity in the food and in the irradiation room will be insignificant.

6.4. MEASUREMENT INSTRUMENTS

Commercial radiation measurement instruments are available and they are applicable for X-ray irradiation facilities even with those operated with a maximum energy of 7.5 MeV.

6.5. CONCLUSIONS

Based upon the above, the radiation safety requirements for machines with 7.5 MeV electrons should not differ from those imposed upon machines with lower energy electrons. These machines should be required to comply with the safety requirements for an industrial facility and should not be required to comply with the regulations for nuclear materials.

7. RECOMMENDATIONS

1. To assess the real need for radiation sources for all types of radiation processing including food irradiation, the IAEA should conduct urgently a global survey of such a need for both isotopic and machine sources in its Member States.

2. The conclusions of this meeting, especially with regard to increasing energy levels of X-ray machines for food irradiation to 7.5 MeV, should be brought to the attention of the International Consultative Group on Food Irradiation (ICGFI) with a view to recommend to the Codex Alimentarius Commission to amend the Codex General Standard for Irradiated Foods at an earliest opportunity.

CONSULTANTS' MEETING ON DEVELOPMENT OF X-RAYS MACHINE FOR FOOD IRRADIATION

Vienna, 16-18 October 1995
Room A-1812

List of Participants

Consultants

Prof. Dieter A.E. **Ehlermann**
Federal Research Center for Nutrition
Engesserstrasse 20
D-76131 Karlsruhe
GERMANY
Tel: 0049 7247 82 3612
Fax: 49-7247-22820
E-mail: dieter.ehlermann@bfe.fzk.de

Dr. Ari **Brynjolfsson**
7 Bridle Path
Wayland, MA 01778
USA
Tel: 508 3581640
Fax: 508 3585518
E-mail:

Dr. M. **Washio**
Sumitomo Heavy Industry Ltd.
63-30 Yuhigaoka
Hiratsuka, Kanagawa 254
JAPAN
Tel: 81463 21 8486
Fax: 81463 21 8990
E-mail:

Dr. Ken-ichi **Mizusawa**
Director/GM Accelerator Division
Nissin-Hlgh Voltage Co. Ltd.
Umezu Takasse-Cho, Ukyo-Ku
Kyoto 615
JAPAN
Tel: 0081758821520
Fax: 0081758648812
E-mail:mizusawa@nhv.nissin.co.jp

Dr. Joe **McKeown**
Director, Science & Technology
AECL Accelerators
436B Hazeldean Road
Kanata, Ontario
CANADA K2L 1T9
Tel: 6138312882
Fax:6138310108
E-mail:

Mr. Robert D. **Jarrett**
Radiation Safety Service Section
Tel: 431 2060 22734
Fax: 431 20607
E-mail:jarrett@nepo1.iaea.or.at

Mr. Vitomir **Markovic**
Indust. Appl. and Chem. Section
Tel: 431 2060 22734
Fax: 431 20607
E-mail:markovic@ripo1.iaea.or.at

Mr. Mordecai **Lapidot**
Indust. Appl. and Chem. Section
Tel: 431 2060 21703
Fax: 431 20607
E-mail:lapidot@ripo1.iaea.or.at

Mr. Kishor **Mehta**
Dosimetry Section
Tel: 431 2060 21660
Fax: 431 20607
E-mail:mehta@ripo1.iaea.or.at

32

52

FedEx *USA Airbill* Express

FedEx Tracking Number **8271 1390 3222**

5PH31

Form I.D. No. 0215

Recipient's Copy

RECIPIENT: PEEL HERE

1 From This portion can be removed for Recipient's records.

Date 5/8/01 FedEx Tracking Number 827113903222

Sender's Name Mark Worth Phone 202 454-5123

Company PUBLIC CITIZEN-CM/CW/ORG

Address 215 PENNSYLVANIA AVE SE

Dept./Floor/Suite/Room

City WASHINGTON State DC ZIP 20003

2 Your Internal Billing Reference

3 To

Recipient's Name Dockets Management Branch Phone 301 827-6860

Company Food and Drug Administration

Address 5630 Fishers Lane

To "HOLD" at FedEx location, print FedEx address. We cannot deliver to P.O. boxes or P.O. ZIP codes.

Room 1061 (HFA-305)

Dept./Floor/Suite/Room

City Rockville State MD ZIP 20852

8271 1390 3222

0174806840

4a Express Package Service *Packages up to 150 lbs.* Delivery commitment may be later in some areas.

- ☐ FedEx Priority Overnight — Next business morning
- ☑ FedEx Standard Overnight — Next business afternoon
- ☐ FedEx First Overnight — Earliest next business morning delivery to select locations
- ☐ FedEx 2Day — Second business day
- ☐ FedEx Express Saver — Third business day
- ☐ NEW FedEx Extra Hours — Later drop-off with next business afternoon delivery for select locations

FedEx Envelope rate not available. Minimum charge: One-pound rate

4b Express Freight Service *Packages over 150 lbs.* Delivery commitment may be later in some areas.

- ☐ FedEx 1Day Freight* — Next business day
- ☐ FedEx 2Day Freight — Second business day
- ☐ FedEx 3Day Freight — Third business day

* Call for Confirmation:

5 Packaging * Declared value limit $500

- ☑ FedEx Envelope*
- ☐ FedEx Pak* — Includes FedEx Small Pak, FedEx Large Pak, and FedEx Sturdy Pak
- ☐ Other Pkg. — Includes FedEx Box, FedEx Tube, and customer pkg.

6 Special Handling Include FedEx address in Section 3.

- ☐ SATURDAY Delivery — Available only for FedEx Priority Overnight and FedEx 2Day to select ZIP codes
- ☐ SUNDAY Delivery — Available only for FedEx Priority Overnight to select ZIP codes
- ☐ HOLD Weekday at FedEx Location — Not available with FedEx First Overnight
- ☐ HOLD Saturday at FedEx Location — Available only for FedEx Priority Overnight and FedEx 2Day to select locations

Does this shipment contain dangerous goods? One box must be checked.

- ☐ No
- ☐ Yes — As per attached Shipper's Declaration
- ☐ Dry Ice — Dry Ice, 9, UN 1845 ____ x ____ kg
- ☐ Cargo Aircraft Only

Dangerous Goods (incl. Dry Ice) cannot be shipped in FedEx packaging or with FedEx Extra Hours service.

7 Payment *Bill to:* Enter FedEx Acct. No. or Credit Card No. below.

- ☑ Sender — Acct. No. in Section 1 will be billed.
- ☐ Recipient
- ☐ Third Party
- ☐ Credit Card
- ☐ Cash/Check
- ☐ Obtain Recip. Acct. No.

Total Packages	Total Weight	Total Charges
1		Credit Card Auth.

†Our liability is limited to $100 unless you declare a higher value. See the *FedEx Service Guide* for details.

8 Release Signature *Sign to authorize delivery without obtaining signature.*

By signing you authorize us to deliver this shipment without obtaining a signature and agree to indemnify and hold us harmless from any resulting claims.

Questions? Visit our Web site at fedex.com or call 1-800-Go-FedEx® (800)463-3339.

406